DASA Report Number 1821

Eleventh Progress Report

on

PROJECT STARDUST

By

Herbert W. Feely
Pierre E. Biscaye
Ben Davidson
Harold Seitz

Prepared for the Defense Atomic Support Agency

July 1, 1966

ISOTOPES, INC.

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ABSTRACT

Work performed on Project Stardust during the two year period, September, 1963 to September, 1965, is reviewed. Gas samples, to be analyzed for carbon-14, have been collected on most Stardust sampling missions since August, 1963 in addition to the filter samples. The RB-57F aircraft has replaced the WU-2 aircraft as the primary vehicle used for the collection of filter samples and gas samples of stratospheric air. The procedures followed in the analysis of the filter samples for products of cosmic-ray activity and the gas samples for carbon-14 are described.

Stratospheric concentrations of radioactive debris, including both fission products and products of neutron activation, were very high in early 1963 as a result of injections during the 1961 and 1962 weapons tests series. During 1963 to 1965 these concentrations decreased as these radionuclides underwent radioactive decay and relatively rapid fallout to the troposphere and to the surface of the earth. As a result, by mid-1965 the fission products had reached levels similar to those found in mid-1961, before the 1958-1961 moratorium on weapons testing was ended. The stratospheric burden of strontium-90, which had reached 6.5 megacuries by the beginning of 1963, decreased to about 2.1 megacuries by mid-1964 and to about 0.9 megacurie by mid-1965. This indicates a stratospheric residence half-time of about ten months for strontium-90, and probably for most particulate debris from the 1961-1962 low altitude weapons tests. The decrease in the manganese-54 burden from about 24 megacuries (corrected for decay to 31 December 1962) to about 3 megacuries in mid-1965 is consistent with this concept. The mean values of the activity ratios Cs^{137}/Sr^{90} (1.56 ± 0.24), $Pu^{239,240}/Sr^{90}$ (0.016 ± 0.005), and Fe^{55}/Mn^{54} (2.4 ± 0.4) in Stardust filter samples were generally independent of time and location of sample collection, indicating that the stratospheric burdens of cesium-137, plutonium-239,240 and iron-55 were also decreasing with a residence half-time of about ten months. On the other hand, carbon-14, present in the stratosphere

ABSTRACT (continued)

as carbon dioxide, showed a decrease in its stratospheric burden from about 36×10^{27} atoms in early 1963 to about 17×10^{27} atoms in early 1965. During these two years the decrease in carbon-14 concentrations approximately corresponded to a stratospheric residence half-time of about 18 months, but the effective residence time was increasing with the passage of time as the fallout rate gradually decreased.

The rate of transfer of particulate radioactive debris, including strontium-90, manganese-54 and yttrium-88, from the northern tropical stratosphere into the stratosphere of the Southern Hemisphere was observed to be seasonally dependent, reaching a maximum during the winter and early spring season of the Southern Hemisphere. A maximum was found in the vertical profile of concentrations of particulate debris in the lower stratosphere during 1963 to 1965, and it showed no tendency to migrate to higher altitudes during that interval. Evidently rates of particle settling in the upper stratosphere exceeded the rates of upward diffusion of the debris from the lower stratosphere, preventing the upward migration of the level of the maximum. The level of the maximum in the vertical profile of bomb-produced $C^{14}O_2$ did appear to occur at higher altitudes as time passed during 1963 to 1965, no doubt as a result of upward diffusion and of removal of the bomb debris from the stratosphere almost entirely at its lower boundary.

Cadmium-109, injected intothe upper atmosphere in July, 1962, was intercepted in the lower stratosphere in the Southern Hemisphere towards the end of 1963, and in the Northern Hemisphere at the beginning of 1964. By 1965 this nuclide was found in similar concentrations in both hemispheres, and appeared to be present in uniform concentrations in the upper stratosphere and down to a height of about 55 thousand feet in the lower stratosphere. The stratospheric burden of this nuclide appeared to decrease from about 70 kilocuries in 1964 to about 50 kilocuries in mid-1965, by which time half of the remaining burden was

present between the tropopause and the 50 mb. level.

SNAP-9A plutonium-238 injected into the upper stratosphere in April, 1964 had entered the lower stratosphere of the Southern Hemisphere by May, 1965 and that of the Northern Hemisphere by December, 1965. Within a few months after entering the lower stratosphere it reached the level of the tropopause. About 2.4 kilocuries of the estimated 17 kilocurie injection were present between the tropopause and the 40 millibar level during late 1965.

Changes in the stratospheric distributions of the cosmic ray products beryllium-7 and phosphorus-32 appeared to accompany changes in the stratospheric circulation during the winter of 1963-1964.

Measurements of lead-210 in samples collected during 1957 to 1959, and of lead-210 and polonium-210 in samples collected during December, 1964 indicated that the Po^{210}/Pb^{210} activity ratio is about 1.0 in the stratosphere, and that there is a layer of maximum lead-210 concentration in the lower stratosphere, at a height of about 45 thousand feet in the polar stratosphere but sloping up to about 60 thousand feet in the tropical stratosphere.

The Stardust numerical model of transfer and rainout of stratospheric radioactive materials has been further developed with the incorporation of a general anisotropic diffusion process. Versions of the model have now been tested in which the principal diffusion axis is along a horizontal surface, along surfaces of constant potential temperature, and approximately along surfaces of constant potential vorticity. Only the last-mentioned of these versions successfully reproduced the observed distributions of radioactivity in the stratosphere.

During mid-1963 and mid-1964 the concentration of artificial carbon-14 from weapons tests reached values of more than 90 percent of the natural carbon-14 level in carbon dioxide collected from ground-level air in northern New Jersey. In mid-1965, however, the highest concentration reached was only 75 percent of the natural level.

Measurements of the distribution of fission products in a series of cores of two New Jersey soils suggested that downward migration of the nuclides flattened the vertical activity profile between August, 1960 and January, 1962, but that the profile had steepened again by October, 1962 and September, 1963 as a result of deposition of fallout from the 1961 and 1962 weapons tests. Deposition rates calculated from these soil samples agree reasonably well with fallout deposition rates calculated from precipitation samples collected at Westwood, New Jersey.

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CHAPTER 1. RECENT PROGRESS ON PROJECT STARDUST

This is the eleventh progress report to be issued during Project Stardust. The last interpretive report issued was the Ninth Quarterly Report on Project Stardust, DASA 1309, dated September 1, 1963. The Tenth Progress Report on Project Stardust, HASL-153, was published by the New York Operations Office of the U. S. Atomic Energy Commission. It contained flight data and results of radiochemical analyses of filter samples collected during 1961 and 1962 under Project Stardust, but included no interpretive sections.

The present report undertakes to review the work which has been performed on Project Stardust during September 1963 - August 1965. An appendix to this report, containing flight data and results of radiochemical analyses of filter samples collected during 1963 under Project Stardust, has been published as report HASL-168 by the U. S. Atomic Energy Commission Health and Safety Laboratory, and the results of carbon-14 analyses performed on stratospheric carbon dioxide samples collected on Stardust sampling missions during August 1963 to April 1965 have been published in report HASL-166. In addition, flight data and results of analysis of Stardust samples for 1964 and 1965 have been published by the Atomic Energy Commission Health and Safety Laboratory as HASL-169 and 176, respectively.

Most of the techniques of sample collection and analysis and of data reduction which are presently being used during Project Stardust are adequately described either in the Final Report on the High Altitude Sampling Program, DASA 1300, or in the Ninth Quarterly Report on Project Stardust, DASA 1309. Therefore, the only techniques described in this report are those not discussed in those two earlier publications: the procedures followed in the analysis of filter samples for strontium-90 and the cosmic ray products beryllium-7,

sodium-22, phosphorus-32 and phosphorus-33, and the procedures followed in analyzing Stardust stratospheric carbon dioxide for carbon-14.

This report also contains a review of recent progress in the study of the stratospheric distributions and stratospheric burdens of radioactive debris from nuclear weapon tests, in the study of the stratospheric distribution of plutonium-238 from the April 1964 SNAP-9A burnup, in the study of the stratospheric distribution of such natural radionuclides as the cosmic ray products and the radon daughter products, in the design and testing of the Stardust numerical model of diffusion and rainout of stratospheric radioactive materials, in the monitoring of carbon-14 concentrations in ground-level air in northern New Jersey, and in measurements of the radioactive fallout in two New Jersey soils.

Purpose and Method of Project Stardust

Project Stardust was undertaken initially to prepare a mathematical model of atmospheric mixing and circulation which could be used to predict stratospheric hold-up and ultimate patterns of deposition on the surface of the earth of radioactive debris injected into the stratosphere by nuclear weapon tests, by burn-up on re-entry of radioactive nuclear power sources for earth satellites, or by other causes. An additional aim of the project, which has now become its primary purpose, is the monitoring of stratospheric concentrations of radioactive debris injected into the stratosphere by past events in order to obtain information needed for the estimation of the potential future hazard from such debris.

To permit the monitoring of stratospheric concentrations of radioactive debris, regular Stardust sampling missions have been flown since June 1961 to collect filter samples of stratospheric air, and, since August 1963,

whole air samples. At Isotopes, Inc. in Westwood, New Jersey the filter samples are analyzed for a series of fission products and neutron activation products, and, at times, for radionuclides produced by natural processes. Aliquots of the whole air samples are measured for their carbon dioxide content using an isotope dilution technique, and the carbon dioxide fractions of the samples are measured for their carbon-14 content.

Information obtained from several atmospheric "tracers" is being used to guide the selection of values for the various meteorological parameters included in the model. Among these tracers are several artificial radionuclides introduced into the stratosphere by nuclear weapon tests, several natural radionuclides found in the stratosphere and troposphere, plutonium-238 from the SNAP-9A power source burn-up in April 1964, and such non-radioactive materials as ozone and water vapor. The results of measurements of atmospheric radioactivity made during Project HASP, Project Stardust, the U. S. Atomic Energy Commission high altitude balloon sampling program, and other measurement programs have been used to determine the atmospheric distribution of the radioactive tracers and the changes which occur in these distributions as a function of time.

The numerical model which is being developed provides an objective method of experimentation in the attempt to simulate the actual processes which affect the movement of radioactive contaminants within the stratosphere. It permits evaluation of the effects in the movement of the contaminant to be expected as a result of the configuration of the source, of the size of the diffusion coefficients, of the possible existence of a general circulation within the stratosphere, of the settling velocity of stratospheric particles and of the possible tropospheric removal mechanisms.

Developments During September 1963 - September 1965

During 1963 to 1965 there has been a continuing decrease in the stratospheric concentrations of radioactive debris from nuclear weapons tests. This decrease has resulted both from the radioactive decay of the radionuclides involved and from the fairly rapid fallout of particulate debris from the stratosphere. In addition, the radioactive debris which has remained in the stratosphere has undergone continued lateral and vertical movement, with the result that the debris in the various regions of the lower stratosphere has become more uniform in concentration and composition. These changes in the stratospheric distribution of radioactive debris have resulted in changes in the sampling and analytical schedules followed in the study of the debris during Project Stardust.

There have been four major changes in the Stardust program during September 1963 to September 1965. The first has involved a decrease in the frequency of sampling missions. Currently the entire Stardust sampling corridor is covered once a month rather than twice a month as during early 1963. In addition the tropospheric sampling which was instituted during 1963 was terminated during 1964. The new, restricted sampling program is adequate to meet the present needs of Project Stardust, especially since some tropospheric samples collected during another program are being made available for analysis under Stardust.

The second change in the program has involved the elimination from the analytical schedule of most of the nuclides which were measured during 1963. By the second half of 1965 only five nuclides, strontium-90, manganese-54, cadmium-109 and plutonium-238, 239, were being analyzed routinely in Stardust filter samples, though carbon-14 was still being analyzed in the whole air

samples collected during the program. All fission products, such as barium-140, strontium-89 and zirconium-95, and activation products, such as yttrium-88 and antimony-124, which have half lives of only a few weeks or months were eliminated during 1964 as their activities became too low to measure accurately. Other longer-lived fission products and activation products, such as cerium-144, promethium-147, cadmium-133m and iron-55, were eliminated during late 1964 or early 1965 as the stratospheric debris became more uniform in composition and, as a result, the measurement of these nuclides ceased to yield useful information.

The third and fourth changes in the program involved the addition of two types of tracer nuclides to the list of radionuclides measured in the filter samples: the cosmic ray products phosphorus-32, phosphorus-33 and sodium-22 and the SNAP-9A fuel nuclide, plutonium-238. At several times in the past some cosmic ray products, and especially beryllium-7, have been measured in Stardust filters, but between late 1963 and early 1965 the most intensive effort yet in the program was made to use these nuclides to elict information on air motions in the stratosphere. Measurements of plutonium-238 were begun in mid-1964 after the announcement was made that a SNAP-9A power source, containing 17 kilocuries of this material, had probably burned up in the stratosphere during reentry after a satellite failed to achieve orbit in April 1964. Plutonium-238 from this source was found in balloon samples during the second half of 1964, and was first found in Stardust samples in mid-1965.

The measurements performed during 1963 to 1965 have yielded useful information on the stratospheric distribution of such potentially hazardous components of radioactive fallout as strontium-90, cesium-137, plutonium-239 and carbon-14. The results of these measurements and the results of measurements of such additional natural and artificial tracer nuclides as the cosmic

ray products, the radon daughter products, lead-210 and polonium-210, the SNAP-9A plutonium-238, the cadmium-109 injected at very high altitudes in July 1962, and the activation products manganese-54, iron-55, antimony-124, yttrium-88 and sodium-22 injected into the lower stratosphere in 1961 - 1962 have all provided additional information which has been or will be of use in determining the nature and rate of the movement of air masses within the stratosphere.

Considerable progress was made during September 1963 to September 1965 in the development and testing of the Stardust numerical model of diffusion and rainout of stratospheric radioactive materials. This two dimensional model, which is capable of simulating diffusion, transport, particle fall velocity and tropospheric rainout, incorporates a general anisotropic diffusion process. The parameters of the model have been varied in an attempt to reproduce quantitatively the known behavior of tungsten-185 injected into the equatorial regions during mid-1958 and, at least qualitatively, the behavior of strontium-90 injected into the stratosphere at high latitudes during 1961 and 1962. Three types of models have been tested; they are characterized respectively by having principal diffusion axes which are (a) along horizontal surfaces, (b) along surfaces of constant potential temperature, and (c) along surfaces of constant potential vorticity. Although all three types can reproduce satisfactorily the latitudinal variation of rainout, only the third can reproduce the observed pattern of concentration in the stratosphere.

Work continued during 1963 to 1965 on two of the ancillary studies performed during Projects HASP and Stardust: the analysis of carbon-14 in ground-level air in New Jersey and the analysis of several fission products in two New Jersey soils. The former is being continued to supplement the studies of carbon-14 in Stardust stratospheric whole air studies, but the

latter has been terminated since its continuation was not justified by the quality of the results obtained.

Future Plans for Project Stardust

During the coming year it is expected that the present rate of sample collection and the present scheme of sample analysis will be continued. The entire Stardust sampling corridor will be sampled approximately once per month. Approximately 20 stratospheric whole air samples will be analyzed for their carbon-14 content each month. Approximately 20 filter samples will be analyzed for their strontium-90, plutonium-238 and plutonium-239 contents each month, and some of these will also be analyzed for manganese-54 and cadmium-109. Additional filter samples will be analyzed for strontium-90 only, and perhaps some will be analysed for lead-210 and polonium-210.

It is not expected that any additional studies of the physical and chemical characteristics of stratospheric particulates, such as were described in the Ninth Quarterly Report, DASA 1309, will be performed under Project Stardust in the future. Neither is it expected that additional work on the numerical model of diffusion and rainout will be performed as part of Project Stardust. It is expected, however, that both of these studies will be continued during the coming year under separate projects sponsored by the Fallout Studies Branch, Division of Biology and Medicine of the U. S. Atomic Energy Commission.

The next interpretive report on the project will be completed about 1 October 1966.

CHAPTER 2. THE STARDUST SAMPLING PROGRAM

The sampling techniques used during Project Stardust were described in the Ninth Quarterly Report, DASA 1309. There have been a few changes in these techniques since the writing of that report, including the initiation of the collection of whole air samples during most Stardust sampling missions the substitution of RB-57F aircraft for WU-2 aircraft in the performance of the missions, and reduction in the frequency and modification of the spacial coverage of the missions.

Collection of Whole Air Samples

Beginning in August 1963 whole air samples have been collected during most sampling missions flown for Project Stardust. Compressed air samples have been collected aboard WU-2, RB-57C and RB-57F aircraft using the sampling system described by Hagemann et al[1]:

> "Whole air samples are obtained aboard aircraft by compressing air into nickel-plated spherical steel bottles about 900 in^3 in volume. Under normal operating conditions each bottle is compressed to about 3,000 psi. The bottles are not evacuated prior to sampling, hence they contain about one atmosphere of ground level air which is added to the sample. No correction is made for this background. The air sample is obtained from the compression section of the jet engine motor and fed to an oil lubricated compressor. The tubing between compressor and bottle is metal with some synthetic rubber connections. Normally four bottles are used to supply the CO_2 for a single sample. There is a positive outward pressure gradient from the bottles from the time of collection to laboratory processing."

All samples are shipped to Airco (the Air Reduction Corporation, Murray Hill, New Jersey) by the units of the U. S. Air Force which collect them. At Airco a small aliquot of the air is drawn off into an evacuated 1,000 ml. flask before the sample is processed. The carbon dioxide is separated from the

remainder of the air sample using standard freeze-out techniques, and with the aliquot of unprocessed air it is shipped to Isotopes, Inc., where the carbon-14 content of the carbon dioxide sample and the carbon dioxide content of the aliquot of unprocessed air are measured.

The Schedule of Collection of Stardust Samples

An attempt was made during the design of the Stardust sampling program to obtain sampling within a meridional corridor. This ideal situation could not be attained in practice, however, and the sampling corridor consists instead of a series of quasi-meridional segments. These segments are essentially contiguous between 75°N and 10°S, but the segments north and south of Australia, which extend from 18°S to 55°S, are far removed in longitude from the others.

The evolution of the Stardust sampling program during 1961 to 1963 was described in the Ninth Quarterly Report. In September 1963 sampling was performed northward and southward from Eielson Air Force Base, Alaska and northward from Davis-Monthan Air Force Base, Arizona at 39 and 43 thousand feet by RB-57C aircraft and at 50, 55, 60, 65 and about 70 thousand feet by WU-2 aircraft. Southward from Davis-Monthan Air Force Base, northward and southward from Albrook Air Force Base, Canal Zone and northward and southward from Laverton R. A. Air Force Base, Australia sampling was performed at 55, 60, 65 and about 70 thousand feet by WU-2 aircraft. Sampling by the WU-2 aircraft was performed once every two weeks while sampling by RB-57C aircraft was performed once every four weeks. Once each month "orbit" missions were flown in the vicinity of 65°N and 36°N by RB-57C, WB-47 or C-130 aircraft. At intervals of once each month or every two months sampling missions were flown from 60°N to 90°N by RB-52 aircraft. In addition, sections of samples collected

for other programs were made available for use on Project Stardust.

Since September 1963 there have been numerous modifications of this program. In February 1964 the alititude coverage of the WU-2 flights northward and southward from Eielson Air Force Base and northward from Davis-Monthan Air Force Base was reduced to four altitudes, 50, 55, 60 and about 66 thousand feet. During the same month a series of "orbit" flights in the vicinity of 38°S at 30, 35, 40 and 45 thousand feet was begun, to be repeated with a frequency of one set every second week. In may 1964 the "orbit" flights in the vicinity of 65°N and 36°N were discontinued. During the same month the flight plans were changed to replace most flights northward from Davis-Monthan and southward from Eielson by flights from Davis-Monthan to Eielson and back again. Similarly, southwaru flights from Davis-Monthan and northward flights from Albrook were largely replaced by flights from Davis-Monthan to Albrook and back again. In June 1964 the frequency of WU-2 sampling missions was decreased from once every two weeks to once every four weeks. The flights northward from Eielson at 39 and 43 thousand feet by RB-57C aircraft were eliminated after October 1964. In November 1964 WU-2 aircraft ceased flying the sampling missions at 50 thousand feet and higher altitudes from Davis-Monthan and Eielson. In their place missions are now flown from Kirtland Air Force Base and Eielson by RB-57F aircraft. The RB-57F aircraft replaced the WU-2 aircraft in missions flown from Albrook Air Force Base in February 1965 and in missions flown from Laverton R. A. Air Force Base in March 1965. Also during March 1965 the "orbit" missions at 38°S, flown by RB-57C and C-130 aircraft, were terminated. Some samples were also received from missions flown during late November-early December 1963 and during April and June 1964 as part of "Project Springfield," but these were not used in Project Stardust. These were flights by RB-57C and

WB-47 aircraft searching for extrusions of stratospheric air into the troposphere.

The flights included in the Stardust sampling program as it exists in late 1965 are summarized in Table 1 and Figure 1.

TABLE 1. FLIGHT TRACKS OF STARDUST MISSIONS DURING LATE 1965

	Northern Limit	Southern Limit	Altitudes (k ft.)
Missions Northward from Eielson A. F. B., Alaska:			
	75°N, 143°W	67°N, 145°W	50, 55, 60, ~65
Missions between Eielson A. F. B., Alaska and Kirtland A. F. B., New Mexico:			
	67°N, 145°W	37°N, 108°W	39, 43, 50, 55, 60, ~65
Missions between Kirtland A. F. B., New Mexico and Albrook A. F. B., Canal Zone:			
	36°N, 103°W	9°N, 80°W	55, 60, 65, ~70
Missions Southward from Albrook A. F. B., Canal Zone:			
	8°N, 80°W	10°S, 82°W	55, 60, 65, ~70
Missions Northward from East Sale R. A. A. F. B., Australia:			
	18°S, 147°E	36°S, 147°E	53, 58, 63, ~70
Missions Southward from East Sale R. A. A. F. B., Australia:			
	38°S, 147°E	55°S, 148°E	53, 58, 63, ~70

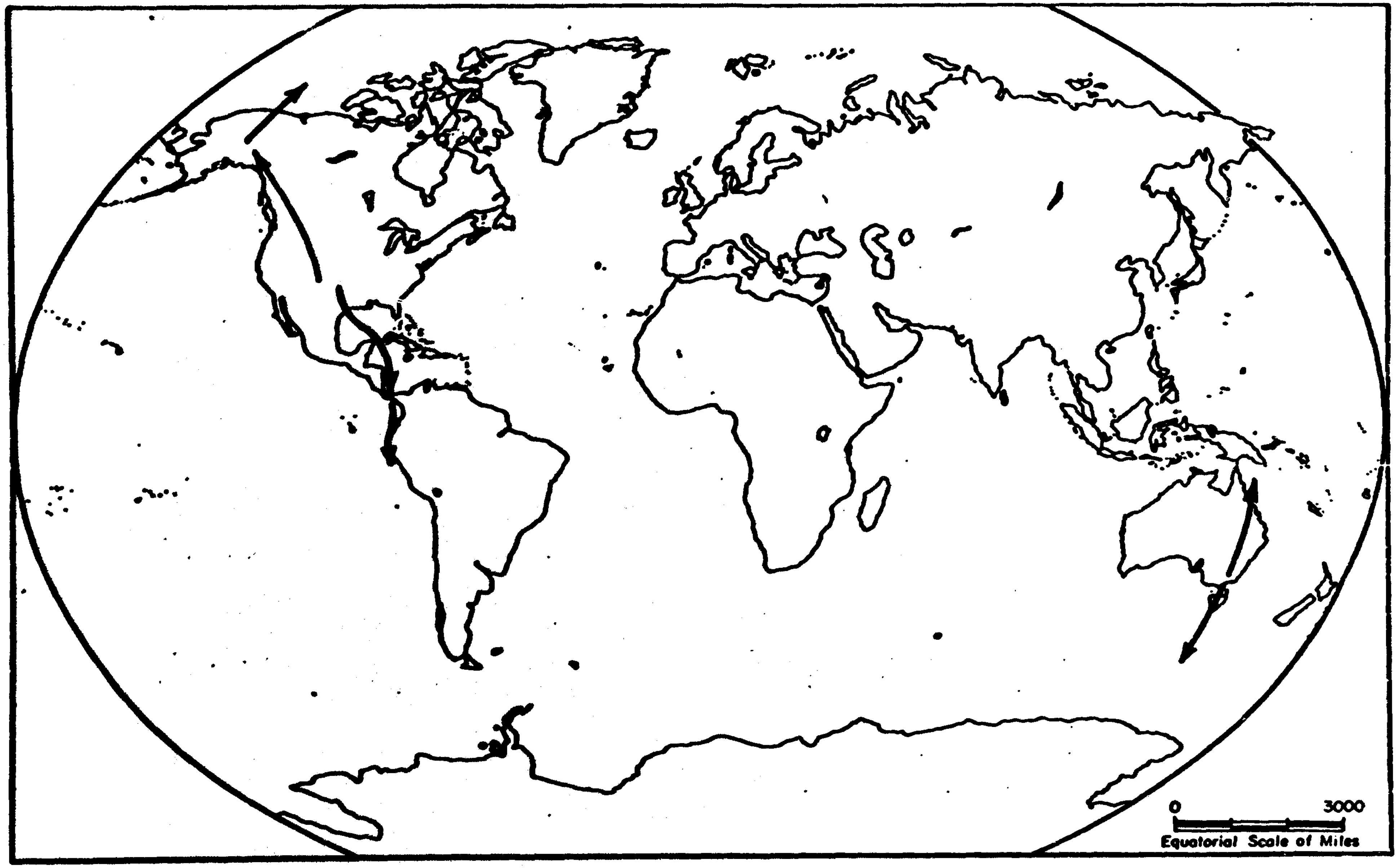


FIGURE 1 · FLIGHT TRACKS OF STAR DUST MISSIONS DURING LATE 1965

CHAPTER 3. THE ANALYSIS OF STARDUST SAMPLES

The purpose and plan of the analysis of filter samples and the actual radiochemical and radiometric procedures followed in measurements of samples collected during the first two years of Project Stardust were described in the Ninth Quarterly Report, DASA 1309. During the two years since that report was prepared some changes have been introduced into the analytical procedures followed, but most of these changes have involved the elimination of nuclides from the scheme of analysis or the combination into a single sequential scheme of several nuclides previously measured as parts of two different sequential schemes. The description of these relatively minor changes would probably not be of value to the reader, so the discussion here will be limited to two major additions to the Stardust analytical program: the analysis of the cosmic ray products, beryllium-7, sodium-22, phosphorus-32 and phosphorus-33 in filter samples, and the analysis of carbon-14 in whole air samples.

The Analysis of Filter Samples for Cosmic Ray Products

Samples which are analyzed for 53 day beryllium-7, 14.3 day phosphorus-32, 25 day phosphorus-33, 2.58 year sodium-22 and 28 year strontium-90 are wet ashed using fuming HNO_3, $HClO_4$, and HF. Aliquots of the resultant solution are taken for determination of phosphorus and sodium before carriers are added to the sample. Beryllium is separated from the solution by precipitation of $Be(OH)_2$, phosphorus is separated as ammonium phosphomolybdate, strontium is separated as $Sr\ CO_3$ and sodium is separated as sodium uranyl acetate.

The beryllium is purified by a series of precipitations which remove several possible radioactive contaminants, and then is extracted into benzene

as an acetyl acetone complex to remove it from any remaining contaminants. It is back-extracted into HCl solution, and is precipitated as $Be(OH)_2$ to be counted for beryllium-7. The phosphorus is converted to $MgNH_4PO_4$ and is passed through two cation exchange columns in succession to remove any radioactive contaminants which are present as cations. It is then reprecipitated as NH_4PMO_4 to eliminate any remaining anionic radioactive contaminants. It is then once more reconverted to $MgNH_4PO_4$ and is heated in a muffle furnace to convert it into $Mg_2P_2O_7$, a form which is most suitable for counting and for yield determination. The sodium, which is precipitated as the uranyl acetate to separate it from possible alkali metal radioactive contaminants, such as cesium-137, is redissolved and is scavenged using several successive $Fe(OH)_3$ precipitations. Any accumulated ammonium salts are then driven off by heating the solution to dryness. The sodium is dissolved and its solution is mixed with 1-butanol which is saturated with HCl to force the precipitation of NaCl, which is counted.

Beryllium-7 is counted on a gamma spectrometer, using the area under the peak due to the 0.48 Mev gamma ray for the assay. Phosphorus-32 and phosphorus-33 are assayed by means of a series of beta counts, using either differential absorption of the 1.71 Mev beta of phosphorus-32 and the 0.25 Mev beta of phosphorus-33 or resolution (by means of a computer program) of the decay curve into a two component system to permit calculation of concentrations of both beta-emitters from the resulting data. The 0.54 Mev position of sodium-22 is also assayed by beta counting the sample, with absorber measurements used to check the energy of the radiation, and, therefore, the radioactive purity of the sample.

Radiochemical and radiometric procedures used to measure strontium-90 were given in the Ninth Quarterly Report. The details of procedures used with

the other nuclides are given below:

A. Ashing Procedure

1. Cut the filter paper which is to be analyzed into small squares and place it in a 600, 800 or 1,000 ml beaker, depending on the quantity of paper used.

2. Add 400 ml of fuming HNO_3 to each sample, cover with a watch glass and allow to digest for 30 minutes. Evaporate the sample down to 25 ml. Care must be taken to reduce the hot plate temperature if any of the reactions become violent.

3. Add an additional 50 ml of fuming HNO_3 to each sample and again evaporate to 25 ml. If any of the solutions are not clear, repeat the treatment with 50 ml fuming HNO_3.

4. Add 50 ml of a 1:1 solution of concentrated $HClO_4$ and concentrated HNO_3 and evaporate to a small volume; do not allow the sample to go to dryness.

5. Transfer the sample to a Teflon beaker using 7N HNO_3 as the transfer agent. Lower the hot plate temperature to about 400° and evaporate the sample to <10 ml.

6. Add 1 ml of concentrated $HClO_4$ and 10 ml of HF to each sample and evaporate down to approximately 5 ml.

7. Repeat step 6 with 10 ml of HF twice. Continue the final evaporation down to drop size. If the sample is not clear at this point, repeat a single HF treatment. If the sample is still not clear, add 5 ml of H_2O_2 and evaporate down to drop size. Add 5 ml of $HClO_4$ and evaporate down to drop size.

8. Transfer the solution into a 25 ml volumetric flask with 0.01N HNO_3.

B. Sequential Separation of Beryllium-7, Phosphorus-32, 33, Sodium-22 and Strontium-90

Because the sample is to be analyzed for sodium-22 the entire sequential separation must be carried out in either teflon or polypropyleneware.

1. Remove four 1 ml aliquots from the dissolved sample in a 25 ml volumetric flask. The sample should be dissolved in approximately 0.01N HNO_3. Dilute to the mark and shake thoroughly before removing aliquots. Two 1 ml aliquots are for natural phosphorus and two 1 ml aliquots are for natural sodium.

2. Pipette 20 ml into two 100 ml teflon beakers.

3. Add the following carriers:

10 ml of 1 mg/ml Beryllium
2 ml of 25 mg/ml Sodium
1 ml of 7.5 mg/ml Phosphorus
1 ml of 30 mg/ml Strontium

Add 1 ml of 2 x 10^4 dpm/ml Strontiumspike

4. Evaporate the sample to a volume of 10 ml and transfer to a clean polypropylene centrifuge tube.

5. Add NH_4OH dropwise until the precipitation of $Be(OH)_2$ is complete (pH = 8). The NH_4OH should be added carefully because if the correct pH for $Be(OH)_2$ is exceeded, phosphorus will also precipitate. Centrifuge, and decant the supernate into a 100 ml teflon beaker.

6. Dissolve the $Be(OH)_2$ precipitate in 5 ml of 6M HNO_3 and dilute to 10 ml with water.

7. Add NH_4OH to the solution from step 6. until $Be(OH)_2$ is completely precipitated. Centrifuge and save this precipitate for beryllium purification. Combine this supernate with the supernate from step 5.

8. In a hot water bath ($\leq 50°C$) add 5 ml ammonium molybdate reagent (saturated) to the combined supernate from steps 5 and 7. Centrifuge and retain this precipitate (ammonium phosphomolybdate) for phosphorus purification.

9. To the supernate from step 8 add NH_4OH to pH >8.5 and add solid $(NH_4)_2CO_3$. Digest in a hot water bath for 5 - 10 minutes, cool and centrifuge. Save the precipitate for strontium purification and retain the supernate for sodium purification.

C. Purifications Procedures for Beryllium-7

1. To the precipitate from the sequential separation, add 10.0 ml 3M NaOH and digest in a hot water bath for <u>not more than 5 minutes</u>. Digestion for longer than 5 minutes will redissolve some of the $La(OH)_3$ precipitate that results from this step. Cool, centrifuge and decant the supernate into 40 ml centrifuge tube. Discard the precipitate $(La(OH_3)$.

2. Neutralize the solution by dropwise addition of conc. HCl, and then make the solution strongly ammoniacal with conc. NH_4OH. The addition of HCl will cause the precipitation of $Be(OH)_2$. But this will re-dissolve when the solution is neutral.

3. Digest in a hot water bath for about 10 minutes, cool, centrifuge $(Be(OH)_2)$ and discard the supernate.

4. Dissolve the $Be(OH)_2$ precipitate in 3 ml of 6M HCl and dilute to 10 ml with water. Add 10 drops each of molybdenum and tellurium scavenging reagent (10 mg Mo, Te/ml) and heat in a hot bath for 15 minutes.

5. To the hot solution add approximately 1 gram of thioacetamide and heat for an additional 15 minutes, filter the solution through a 9.0 cm. Whatman #42 filter paper in a 60°-2" glass funnel.

Collect the filtrate in a clean 125 ml Erlenmeyer flask. The thioacetamide will precipitate both molybdenum and tellurium sulfides. In order to get complete separation from the beryllium, they should be filtered while hot.

6. Evaporate the filtrate to a volume of approximately 3 ml, cool and dilute to a volume of 15 ml with water.

7. Add 2.0 ml of "acetate buffer" and 2.0 ml of 10% EDTA solution. Adjust the pH to 5.5-6.0 by dropwise addition of NH_4OH. Transfer the solution to a 60 ml cylindrical sep. funnel using water as a transfer agent. Add 2.0 ml of acetylacetone and stir on a mechanical stirrer for several minutes.

8. Add 7.0 ml of benzene and stir mechanically for <u>two</u> minutes. Withdraw the aqueous layer into a clean 40 ml centrifuge tube and transfer the organic phase into a second clean 40 ml centrifuge tube.

9. Adjust the pH of the <u>aqueous</u> solution to 5.5-6.0 and transfer the solution back into the sep. funnel.

10. Repeat set 8 and combine the benzene fractions in the sep. funnel.

11. Wash the benzene fraction <u>twice</u> with 10 ml of 10% EDTA.

12. Add to the benzene fraction 7.0 ml of 6M HCl and stir mechanically for several minutes and withdraw the aqueous layer (HCl) into a 125 ml Erlenmeyer flask.

13. Repeat step 12 combining the HCl fractions in the Erlenmeyer flask.

14. Evaporate the HCl fractions almost to dryness. Add 5.0 ml conc. HNO_3 and evaporate to dryness.

15. Dissolve the residue, $Be(NO_3)_2$, in 2.0 ml of 6M HCl (heat if necessary to get all the residue into solution), and dilute with 5 ml of water. Transfer the solution to a clean 40 ml centrifuge tube. Wash the

flask with three 3 ml aliquots of water and add these washings to the centrifuge tube.

16. Make the solution basic with NH_4OH. Centrifuge and discard the supernate. Wash the $Be(OH)_2$ precipitate twice with 5 ml of water, centrifuge and discard the washings.

17. Dissolve the precipitate in a minimum of conc. HCl and transfer the solution to a 10 ml test tube. Make the solution basic with conc. NH_4OH to re-precipitate the $Be(OH)_2$. Centrifuge and discard the supernate.

18. The $Be(OH)_2$ precipitate is counted in this form (damp precipitate) in the 10 ml test tube in a well-type counter (described below) and beryllium is yielded colorometrically after radioassay. The beryllium colorometric procedure is detailed on pp 97-98 of DASA 1309.

D. Purification Procedures for Phosphorus-32, 33:

1. Dissolve the NH_4PMO_4 precipitate from step 8 of the sequential separation in 1.0 ml conc. NH_4OH. If the precipitate does not completely dissolve, centrifuge and decant the supernate into a 40 ml centrifuge tube and discard the remaining precipitate. Add 2 ml of 1N citric acid solution.

2. Slowly add 10 ml saturated magnesia mixture while stirring, and add conc. NH_4OH dropwise until the solution is just alkaline (pH~7); then add 10 more drops of NH_4OH.

3. Stir for 1 minute after formation of the precipitate, and then add 4 ml of conc. NH_4OH. Allow the mixture to stand for at leat 4 hours with additonal stirring.

4. Prepare two cation exchange columns using 10 mm O.D. glass tubing and 10 ml of wet 50 - 100 mesh Dowex AG-50WX 8 cation exchange resin.

5. Condition the columns by passing 150 ml of 1.03N HCl through each and do not let the resin run dry.

6. Centrifuge the precipitate ($MgNH_4PO_4$) from step 3 and discard the supernate.

7. Dissolve the $MgNH_4PO_4$ in 1 ml (pipetted) of 3N HCl and add 99 ml of water. If the solution is not completely clear, heat on a hot plate until it is.

8. Pass the solution from step 7 through one of the conditioned exchange columns at a rate of 1 drop every 5 seconds and collect the efficient in a 150 ml beaker.

9. Pass the efficient through the second conditioned exchange column at a rate of one drop every 5 seconds and collect the effluent in a 150 ml beaker.

10. By pipette add 1 ml of conc. $HClO_4$ and evaporate the effluent until the appearance of fumes of $HClO_4$. Transfer the solution to a 40 ml centrifuge tube with 20 ml of water and by pipette add 4 ml conc. HNO_3.

11. Add 5 ml of saturated ammonium molybdate reagent, several drops of aerosol wetting agent and heat for 5 minutes in a hot water bath ($\leq$ 50°C). Cool, centrifuge and discard the supernate.

12. Dissolve the HN_4PMoO_4 precipitate in 1 ml (pipetted) conc. NH_4OH. Again, if the precipitate does not completely dissolve, centrifuge and decant the supernate into a 40 ml centrifuge tube. Add 2 ml 1N citric acid solution.

13. Repeat step 2 with the magnesia mixture.

14. Repeat step 3.

15. Filter the precipitate onto a Whatman #42 filter disc, wash the precipitate with three 5 ml portions of 1:20 NH_4OH and finally with an-

hydrous "Anhydrol." Dry in an oven at 110°C for 10 minutes and transfer filter paper and precipitate to a porcelain crucible.

16. Char the paper in the crucible using a Bunsen burner. Ash the sample in a muffle furnace at 1050°C for one hour. Cool, transfer the precipitate to a 40 ml centrifuge tube using anhydrous "Anhydrous" as a transfer agent.

17. Carefully grind the precipitate with a glass stirring rod, taking care that no glass is broken off of the stirring rod when grinding. Slurry and filter onto a previously washed and weighed Whatman #42 filter disc using anhydrous "Anhydrol" as a transfer agent. Dry in an oven at 110°C for 15 minutes, cool in a desiccator, weigh, and mount on a brass planchet. A chemical yield correction must be made due to natural phosphorus in the filter. An aliquot of the original sample is taken and measured colorometrically for such phosphorus.

E. Purification Procedures for Sodium-22:

In the sodium-22 purification procedure demineralized water, plastic tubes, teflon beakers and stirrers are to be used exclusively.

1. Transfer the supernate containing 20 mg of sodium carrier to a 100 ml teflon beaker and evaporate to dryness.

2. Cool and add 1 ml of 6M NH_4OAc and 75 ml of sodium precipitating reagent (45g uranyl acetate, 300 g magnesium acetate, 60 ml acetic acid).

3. Stir on a mechanical stirrer for 20 minutes; then centrifuge portion-wise into a plastic centrifuge tube and discard the supernate.

4. Wash the precipitate once with 20 ml of sodium precipitating reagent and once with 20 ml of "sodium wash solution" (35 ml acetic acid, 405 ml anhydrous ethyl acetate, 460 ml 100% ethanol).

5. Dissolve the precipitate in a minimum of concentrated HCl and dilute to 15 ml with water.

6. Add 3 drops of iron carrier (~10 mg/ml) and precipitate the iron with concentrated NH_4OH. Centrifuge and decant the supernate into a clean plastic centrifuge tube. Wash the precipitate with 5 ml deionized water; centrifuge, decant, wash and add to the supernate above. Discard the iron precipitate.

7. Perform two additional iron scavenges. Dissolve the iron from each scavenge with concentrated HCl and then precipitate it with concentrated NH_4OH. Centrifuge, wash with deionized water, centrifuge and add decanted wash to supernate.

8. Transfer the supernate and wash from the final iron scavenge to a teflon beaker and evaporate to dryness.

9. Cool and add 4 ml of concentrated HNO_3 and 1 ml of concentrated HCl and evaporate to dryness under very low heat.

10. Repeat step 9 three times washing down the walls of the beaker with water each time.

11. Add 5 ml of concentrated HCl and evaporate to dryness.

12. Dissolve the precipitate in a minimum of water and transfer to a plastic centrifuge tube. Wash the walls of the beaker with a minimum of water and add this to the tube.

13. Place the plastic tubes in an ice bath and add 15 ml of 1-Butanol saturated with HCl gas. Stir vigorously, and let stand until the ice bath comes to room temperature. To prepare 1-Butanol saturated with HCl, transfer 1/2 pint of 1-Butanol to another bottle. Place the remaining 1/2 pint in an ice salt bath and pass HCl gas through the solution for one hour.

Keep this solution ice cold until ready for use. CAUTION A closed bottle of the saturated solution is explosive at room temperature.

14. Centrifuge and discard the supernate. Add 15 ml of 100% EtOH and stir vigorously. Filter onto a tared #42 Whatman filter paper using 100% EtOH only as a transfer solution.

15. Dry at 100°C for 30 minutes. Cool in a dessicator, weigh and record data as NaCl. Mount for sodium-22 beta counting.

F. Radioassay Procedures for Beryllium-7:

The 0.48 mev γ radiation (53 day half life) is measured to obtain the abundance of beryllium-7 in Stardust samples.

The beryllium is in the form of a moist $Be(OH)_2$ precipitate in a 10 ml test tube following the purification procedure. It is counted in this form in the test tube in a NaI(Tl) well-type crystal (1-3/4" x 2"). The height of the precipitate in the tube is measured and the counting efficiency is determined from a previously constructed calibration curve on which efficiency is plotted against height. All samples are assayed twice, and normally excellent agreement is obtained between the first and second counts. Usual counting periods are approximately an hour in length. Five per cent of the samples are followed for decay to check on half-life and sample purity. A branching ratio of 11.00% for the gamma ray is used in computing beryllium-7 content.

G. Radioassay Producedures for Phosphorus-32, 33:

These β-emitting nuclides are counted using low-level, gas-flow detectors in the geiger region. Phosphorus-32 emits a 1.71 mev β (half-life = 14.3d) and phosphorus-33, a 0.25 mev β (half-life = 25d). The phosphorus precipitate from the purification procedure, counted on a brass planchette is first counted without absorbers and with a minimum distance between sample

and detectors for both phosphorus isotopes. Then the sample is recounted in the same geometry but with a 71.6 mg/cm^2 aluminum absorber between it and the detectors to cut out all phosphorus-33 betas. The sample is counted in this configuration three to six times to follow the decay of 14.3 day phosphorus-32 and the disintegration rate of that nuclide is calculated. The sample is counted without the absorber for the same length of time, and the calculated phosphorus-32 disintegration rate is subtracted from the total disintegration rate of the sample without absorber to obtain the disintegration rate for phosphorus-33. Each count is normally about 7 hours in duration.

H. Radioassay Procedures for Sodium-22:

This nuclide, in the form of the final sodium-chloride precipitate from the purification procedure, is counted with a low level gas flow detector in the geiger region. Sodium-22 emits 0.54 Mev positron radiation, and samples are counted for from two hundred to four hundred minutes. Sample purity is checked by means of a 14.6 mg/cm^2 aluminum absorber.

The Measurement of Carbon-14 in Whole Air Samples

The carbon dioxide samples received from the Air Reduction Company are sufficiently pure that they can be counted after a minimum of additional purification, which is accomplished by passing the carbon dioxide through charcoal at room temperatures. Occasional tests have been made to assure that no isotopic fractionation of the carbon dioxide occurs during this final purification step. These tests have involved measurements of C^{13}/C^{12} ratios and carbon-14 concentrations in carbon dioxide samples before and after passage through the charcoal.

The purified samples are introduced into internal gas proportional counters of copper and with 1 ml tungsten anode center wires. The active volume of each counter is approximately 250 ml. The counters are shielded by 16 inches of steel and 4 inches of paraffin to remove the soft cosmic ray component of the background activity. The hard cosmic ray component, cosmic ray mesons, are removed by an anticoincidence technique using an annular multi-anode anticoincidence guard counter surrounding each sample counter. This total shielding array reduces the background of the 250 ml counters to about 2.2 cpm.

Samples are counted sufficiently long to insure that the statistical counting errors are 5 percent or less. The counters have been calibrated using standard samples of $C^{14}O_2$ supplied by Argonne National Laboratory. The efficiency of the counters for the 15.6 kev. beta of carbon-14 is 92 percent.

The data obtained from the carbon-14 measurements yield only the carbon-14 concentrations of the carbon dioxide in the whole air samples obtained from the aircraft sampling systems. As has been pointed out by

Hagemann et al[1], some carbon dioxide may be added to the whole air samples by oxidation of compressor lubricating oil in the aircraft sampling systems. Because this oil is derived from fossil fuels, the carbon dioxide produced from it should add no carbon-14 to the sample, but rather would act to dilute the carbon-14 already in the air. This dilution is corrected for, in Stardust samples, by measuring the actual carbon dioxide concentrations in the whole air samples and then calculating the carbon-14 concentration in the whole air samples. As long as no significant increase in the volume of the air samples has been produced by the contaminants from the compressor, this carbon-14 concentration may be taken as equal to that in the uncontaminated air sample.

For 76 of the first Stardust samples collected during 1963 we must rely on infrared measurements of carbon dioxide abundances in the whole air samples made at Air Reduction Corporation during the processing of the samples. For subsequent samples, however, measurements of carbon dioxide abundance have been made at Isotopes, Inc. on the air aliquots supplied with the separated carbon dioxide samples. An isotope dilution technique utilizing mass spectrometer measurements has been used. The measurements of abundance made are precise to within $\pm$ 2%. The advantage of this method over the infrared measurements is that its accuracy is not affected by the presence of traces of contaminants such as methane or carbon monoxide in the whole air sample.

An accurately measured aliquot of the whole air sample is taken and to it is added an accurately measured aliquot of a standard "spike" sample enriched in $C^{13}O_2$. The mixture is equilibrated by two successive freeze-outs in a cold finger at liquid nitrogen temeperatures, and gases volatile at these temperatures, such as oxygen, nitrogen and carbon monoxide, are pumped off of the frozen sample.

The C^{13}/C^{12} ratio in the mixture is measured using a Nuclides Corporation RMS-15 multiple collector ratio mass spectrometer. The ratio measured is that of ions of mass 45 ($C^{13}O_2$) to ions of mass 44 ($C^{12}O_2$). At least ten successive determinations of the ratio in the mixture of sample and spike are alternated with at least ten determinations the ratio in a standard sample. The computed average ratio in the mixture is precise to within ± 0.1%. Since the quantity of carbon-13 contributed to the mixture of the whole air sample is negligible compared to the quantity contributed to the mixture by the enriched carbon-13 spike sample, the amount of carbon-12 contributed by the whole air sample can be calculated from the known C^{13}/C^{12} ratio in the spike and the measured C^{13}/C^{12} ratio in the mixture. Since the aliquot of air which contributed this carbon-12 was measured accurately, this information yields the carbon-12 content, and therefore the carbon dioxide content, of the air aliquot.

The values of carbon-14 abundance in the whole air samples calculated from the results of the carbon-14 and carbon-dioxide measurements should generally be accurate to within ± 5%. They are expressed in atoms of carbon-14 per gram of air. To correct these samples for the presence of carbon-14 produced by cosmic ray activity we have assumed, following Hagemann et al, that each sample contains 74×10^5 atoms/g of natural carbon-14, and have subtracted this amount from each result to obtain the concentration of "carbon-14 excess."

CHAPTER 4. THE STRATOSPHERIC DISTRIBUTION OF RADIOACTIVE DEBRIS FROM NUCLEAR WEAPONS TESTS

The series of tests of nuclear weapons which were performed in the atmosphere during September 1961 - December 1962 resulted in the injection of large quantities of radioactive debris into the stratosphere. This debris consisted mainly of fission products, such as molybdenum-99, zirconium-95 and strontium-90, but it also contained unusually high concentrations of certain products of neutron activation, such as iron-55, manganese-54 and antimony-124, which were produced in especially large quantities by some of the very high yield devices tested by the U.S.S.R. in late 1961 and late 1962.

All of the radioactive debris from the two test series performed by the U.S.S.R. was injected into the stratosphere of the Northern Hemisphere. As a result, the concentrations of radioactivity encountered in this region by Stardust sampling missions during early 1963 were generally very high, higher than at any equivalent time during Projects HASP and Stardust. During 1963, 1964 and 1965, however, these concentrations decreased rapidly, both as a result of radioactive decay and as a result of fallout of debris to the troposphere.

In the succeeding sections of this chapter we will discuss the distribution within the Stardust sampling corridor of total beta activity, of strontium-90 and other fission products, of manganese-54 and other activation products produced by low altitude tests during 1961 and 1962, of cadmium-109 produced by the Starfish Prime high altitude event, and of carbon-14.

The Stratospheric Distribution of Total Beta Activity

The mean monthly distributions of total beta activity in the Stardust sampling corridor during September 1963 to August 1965 are plotted in Figures

2 to 13. In September 1963 the total beta activities encountered in the northern polar stratosphere were as much as ten times as high as those encountered in the southern polar stratosphere. By August 1965 the activities in the Northern Hemisphere were at most about twice as high as those found in the Southern Hemisphere. The combination of radioactive decay and fallout had reduced the activities in the northern polar stratosphere by about a factor of 40, and those in the southern polar stratosphere by about a factor of 8, over the course of two years.

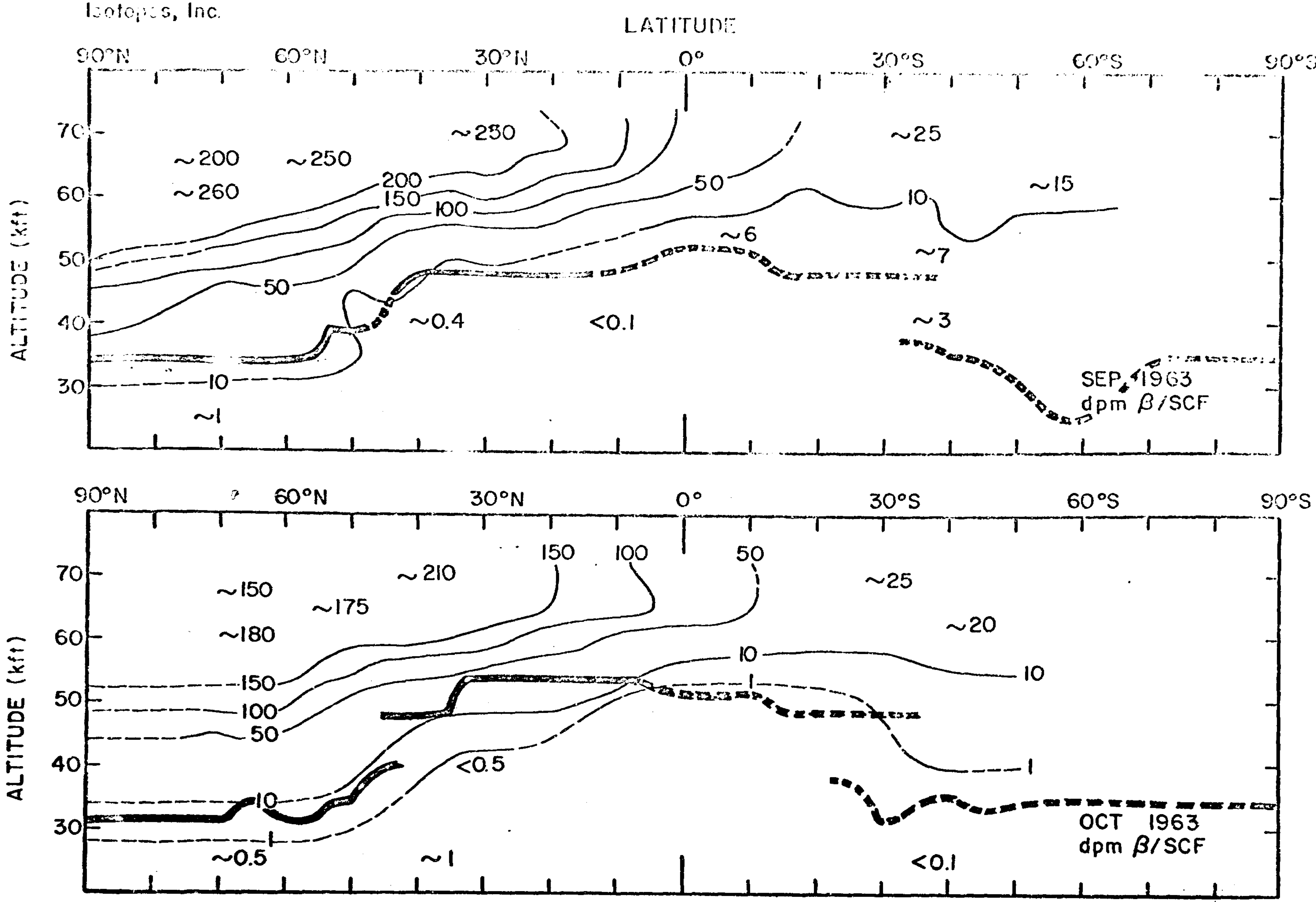


FIGURE 2 · THE MEAN DISTRIBUTION OF TOTAL BETA ACTIVITY DURING SEPTEMBER AND OCTOBER 1963

Isotopes, Inc.

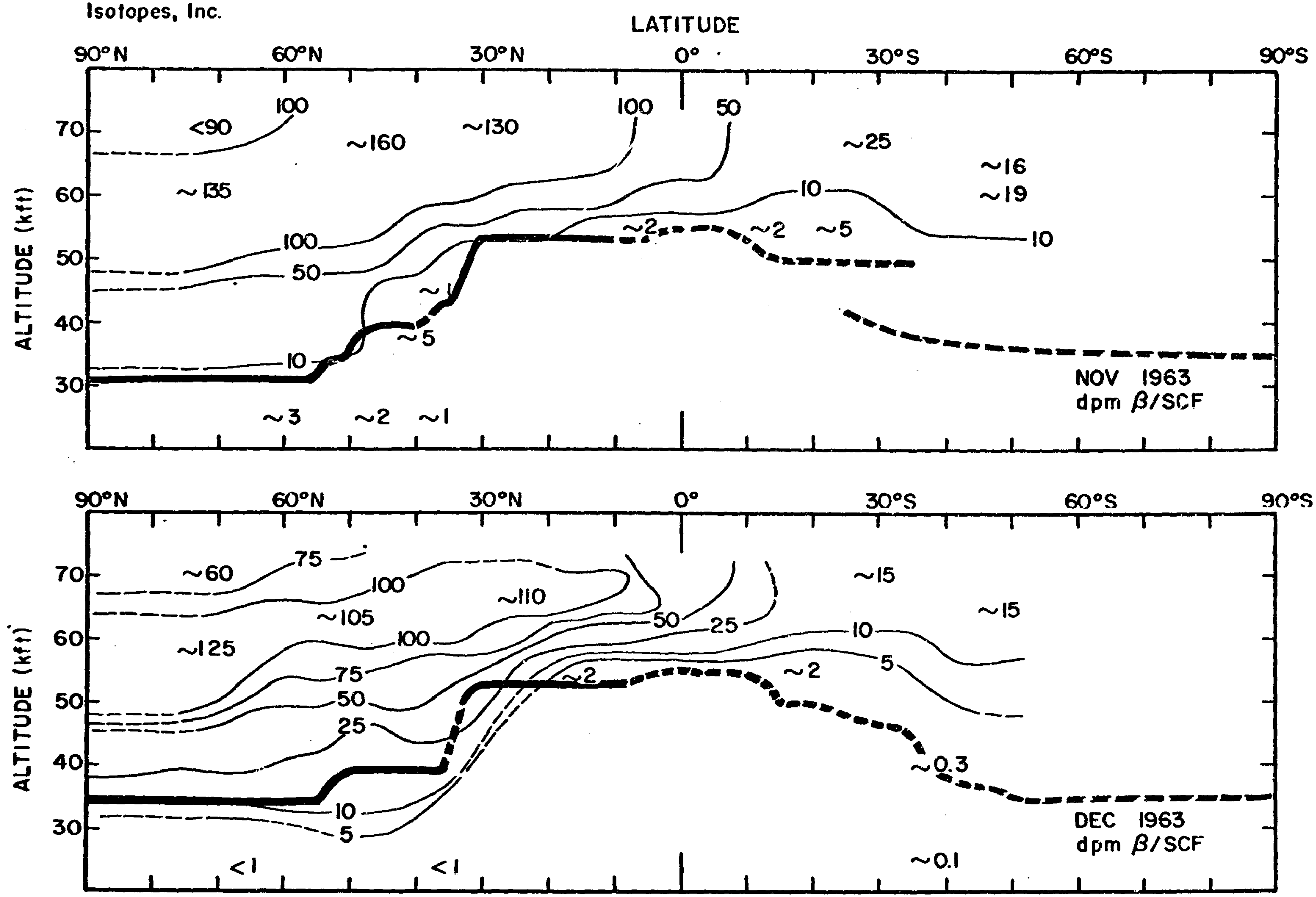
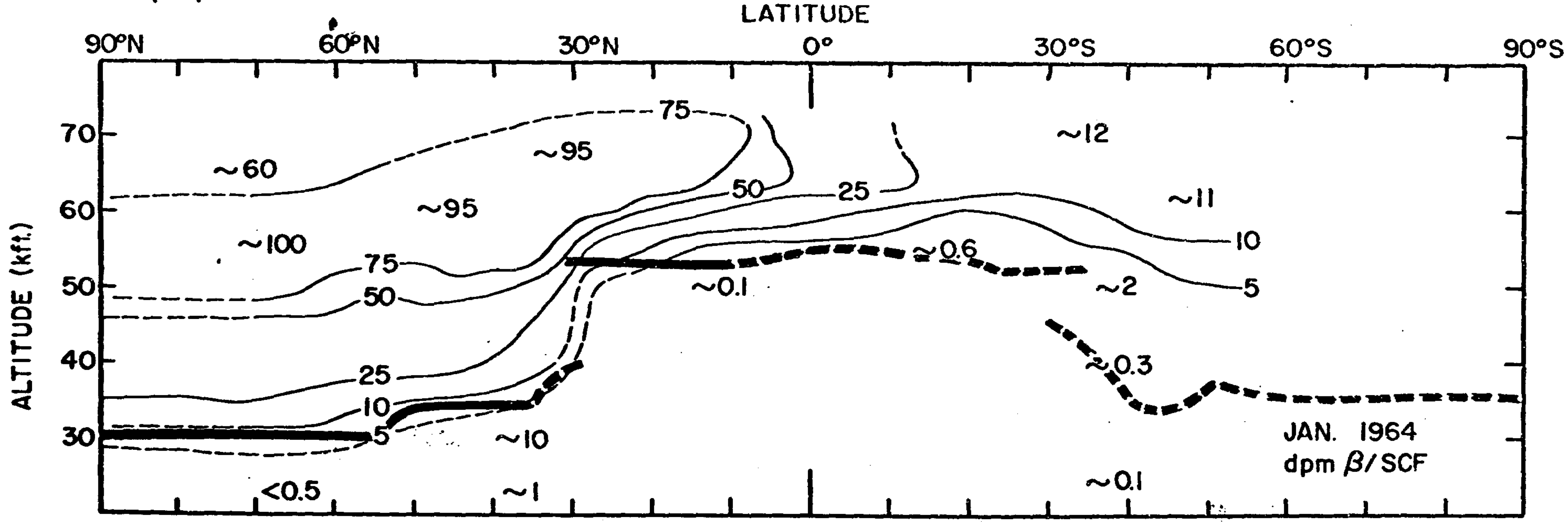


FIGURE 3 · THE MEAN DISTRIBUTION OF TOTAL BETA ACTIVITY DURING NOVEMBER AND DECEMBER 1963

Isotopes, Inc.



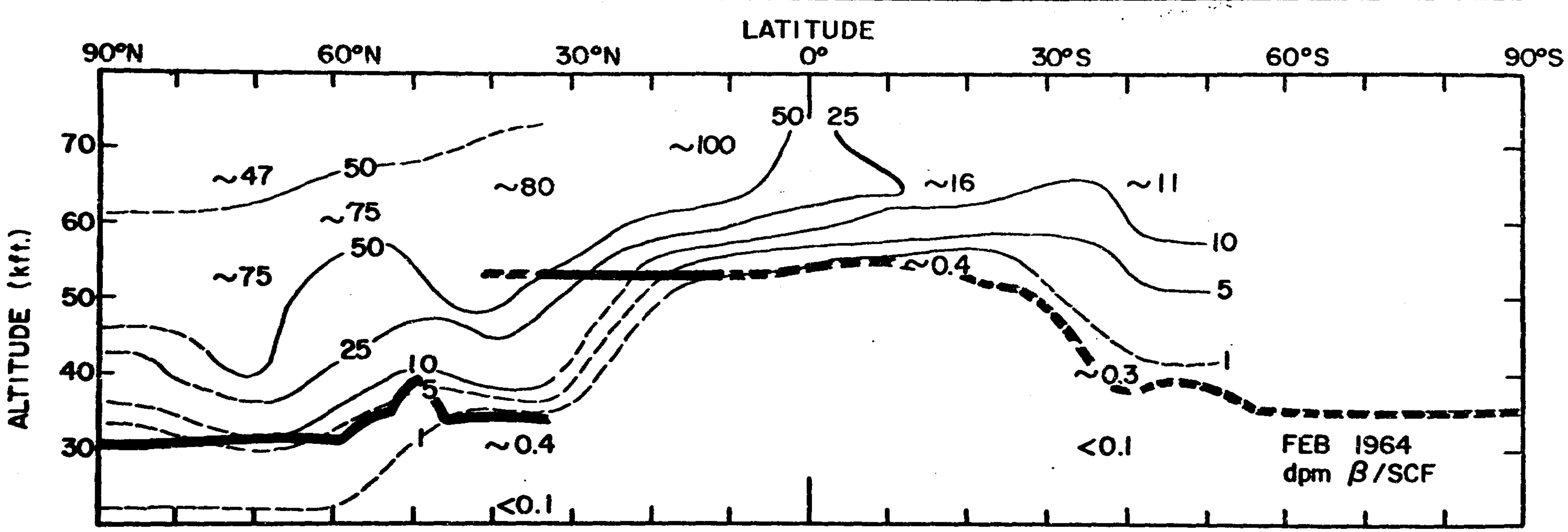


FIGURE 4 · THE MEAN DISTRIBUTION OF TOTAL BETA ACTIVITY DURING JANUARY AND FEBRUARY 1964

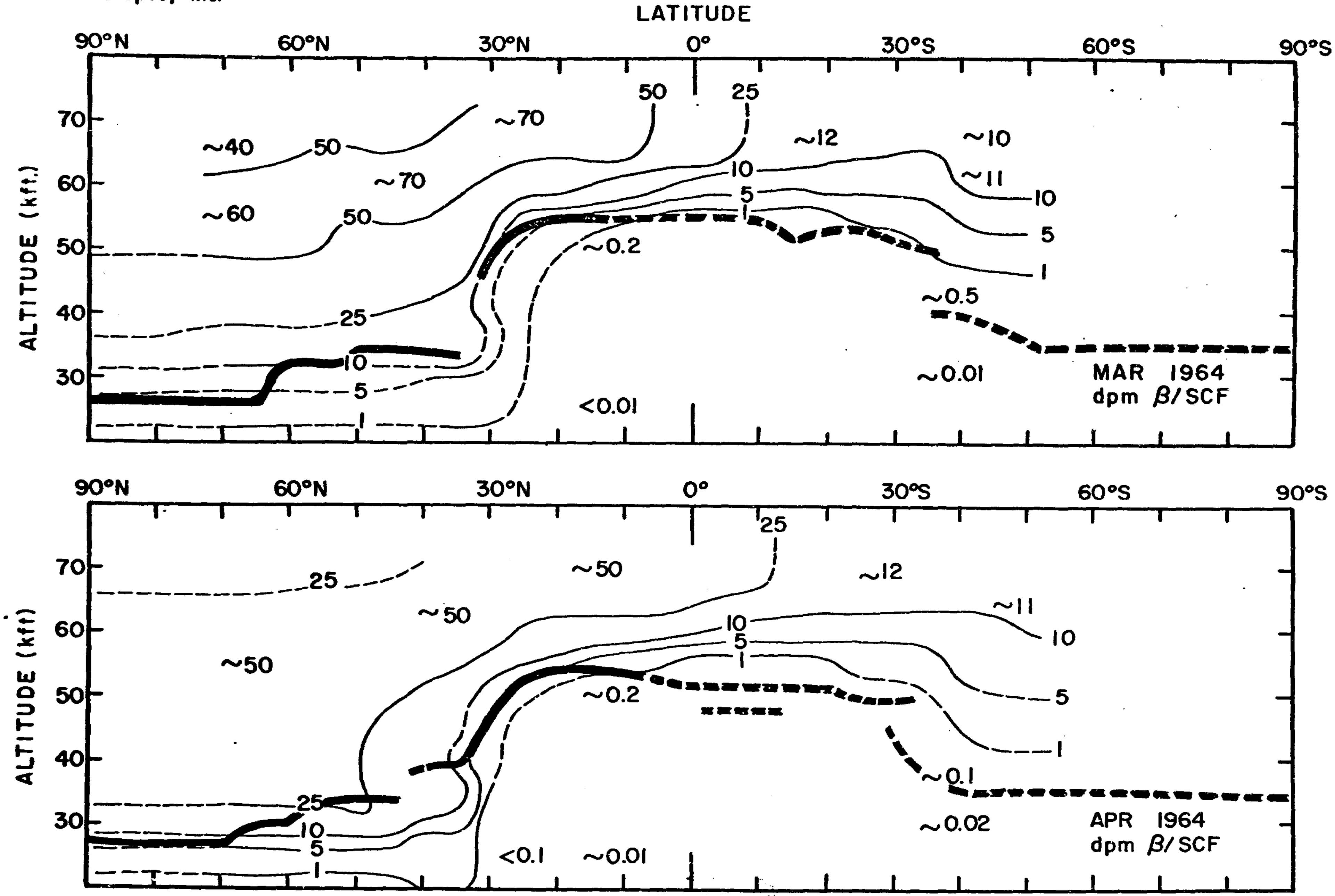


FIGURE 5· THE MEAN DISTRIBUTION OF TOTAL BETA ACTIVITY DURING MARCH AND APRIL 1964

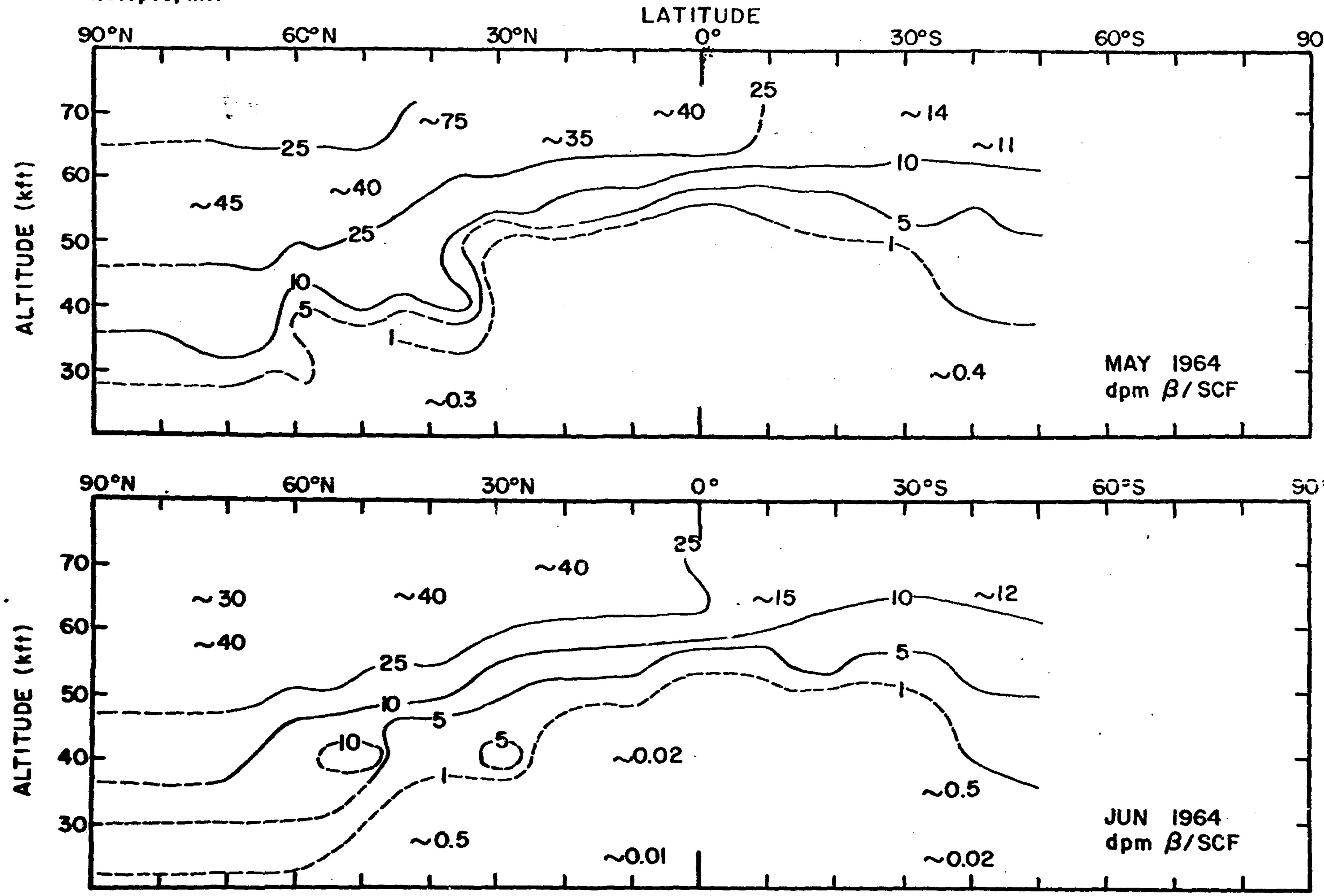


FIGURE 6 · MEAN DISTRIBUTION OF TOTAL BETA ACTIVITY DURING MAY AND JUNE 1964

Isotopes, Inc.

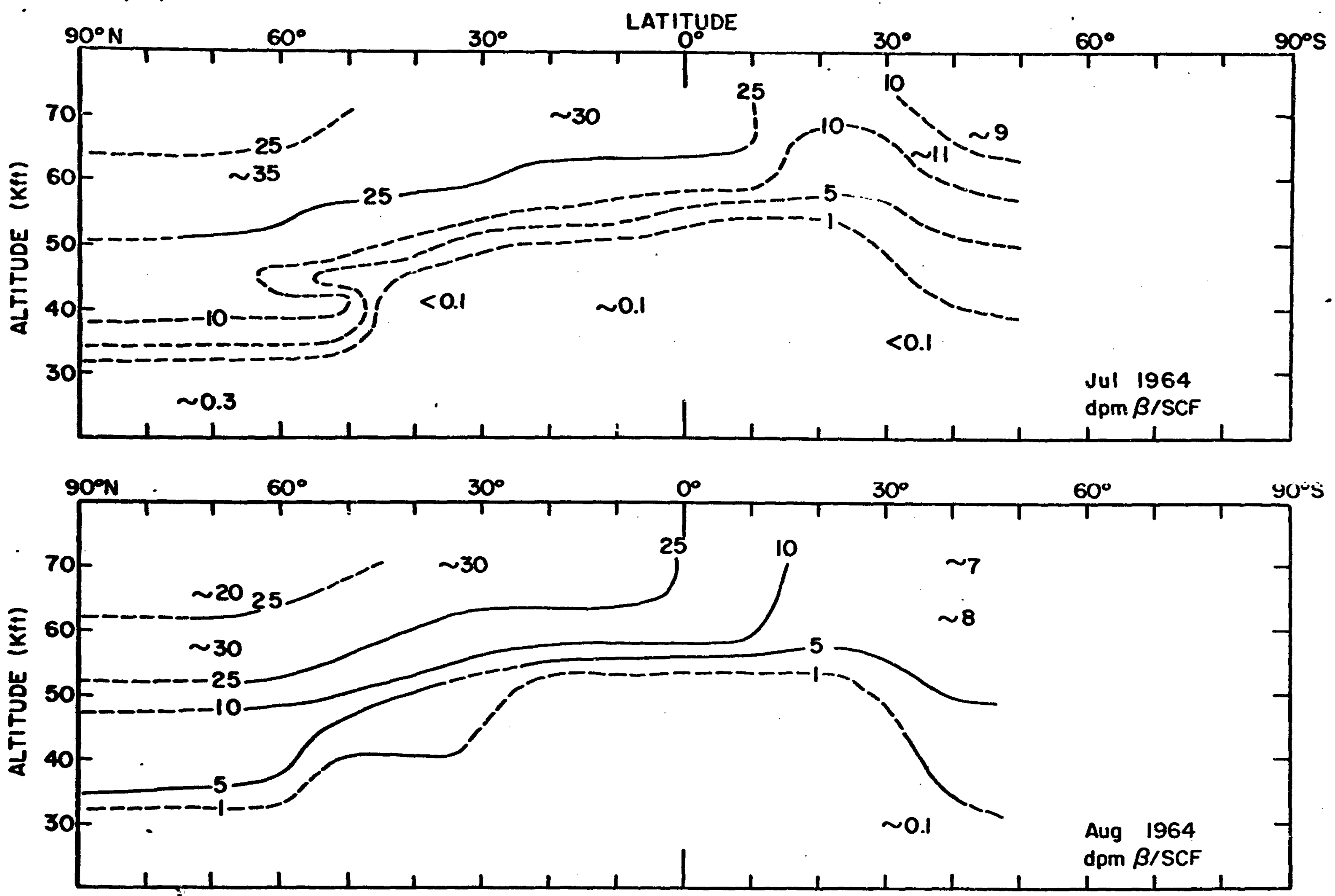


FIGURE 7· THE MEAN DISTRIBUTION OF TOTAL BETA ACTIVITY DURING JULY AND AUGUST 1964

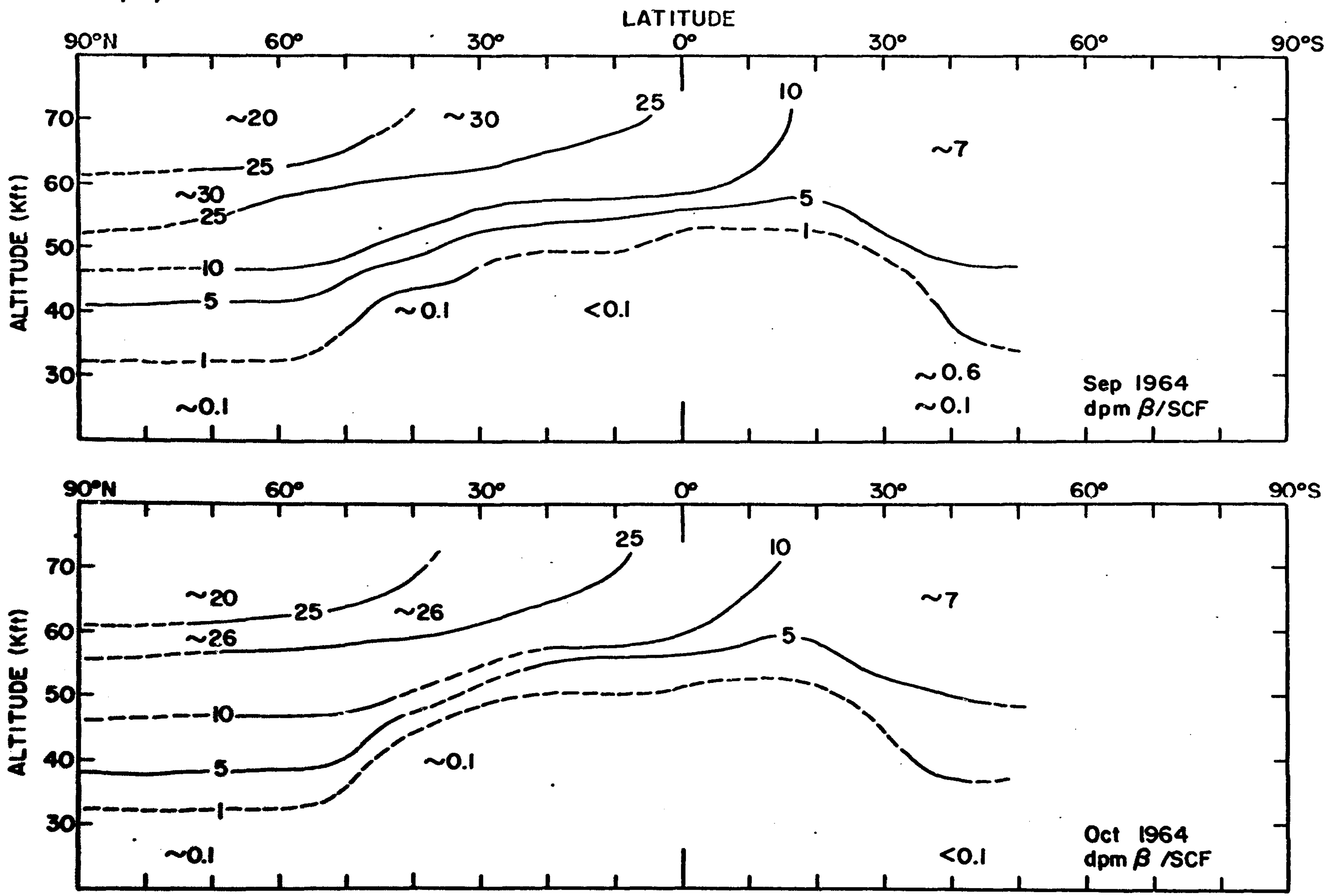


FIGURE 8 · THE MEAN DISTRIBUTION OF TOTAL BETA ACTIVITY DURING SEPTEMBER AND OCTOBER 1964

Isotopes, Inc.

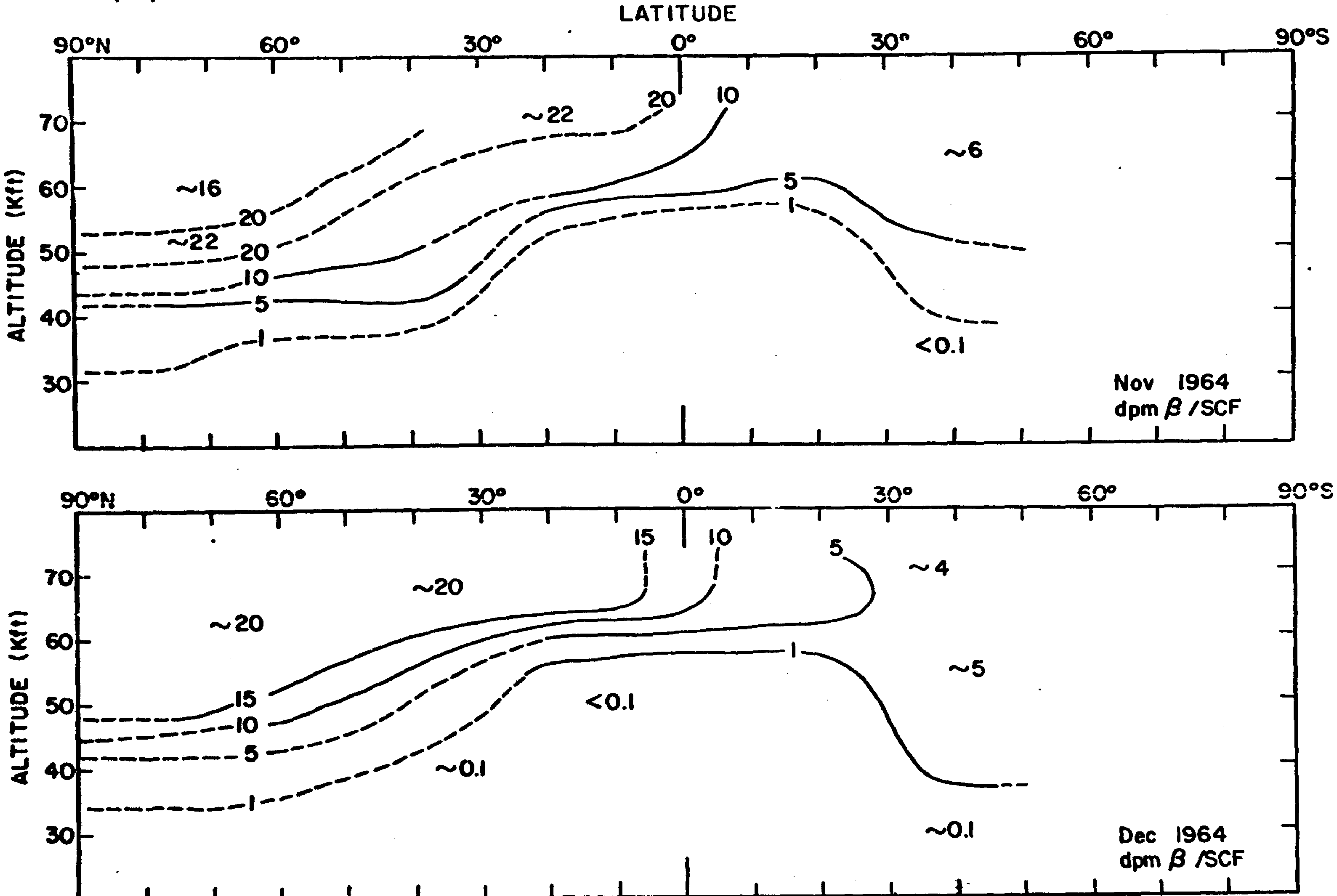


FIGURE 9 · THE MEAN DISTRIBUTION OF TOTAL BETA ACTIVITY DURING NOVEMBER AND DECEMBER 1964

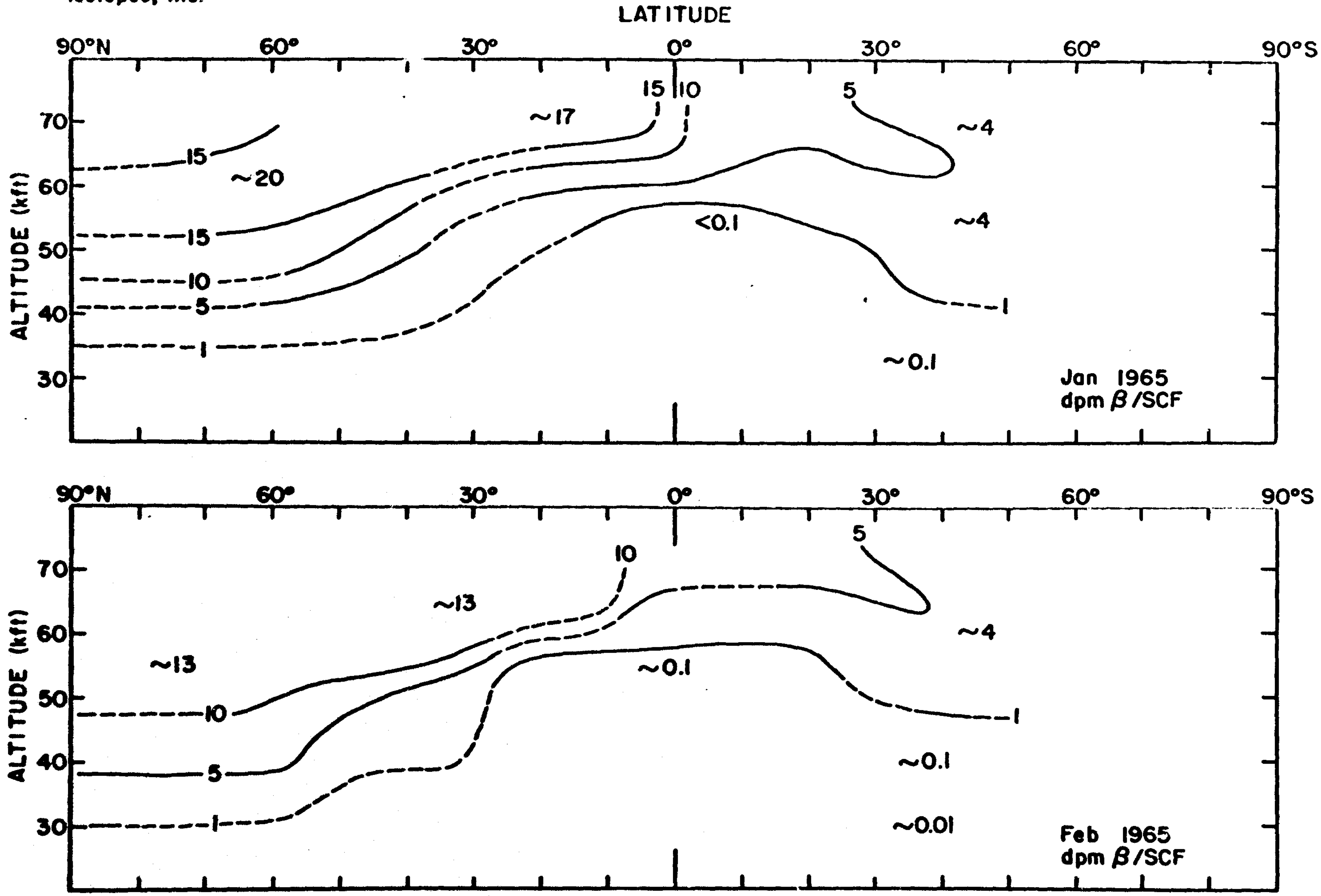


FIGURE 10 · THE MEAN DISTRIBUTION OF TOTAL BETA ACTIVITY DURING JANUARY AND FEBRUARY 1965

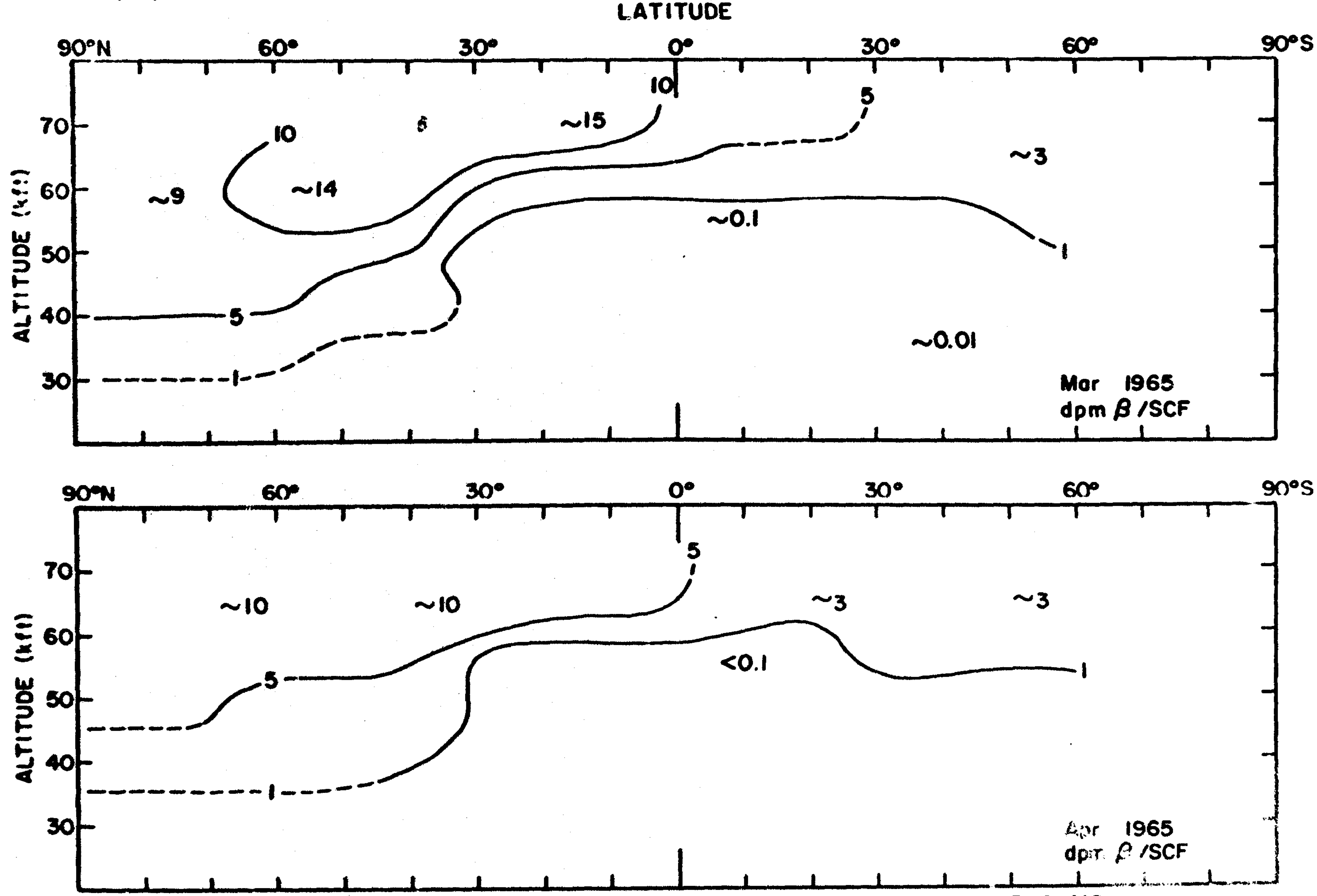


FIGURE II · THE MEAN DISTRIBUTION OF TOTAL BETA ACTIVITY DURING MARCH AND APRIL 1965

Isotopes, Inc.

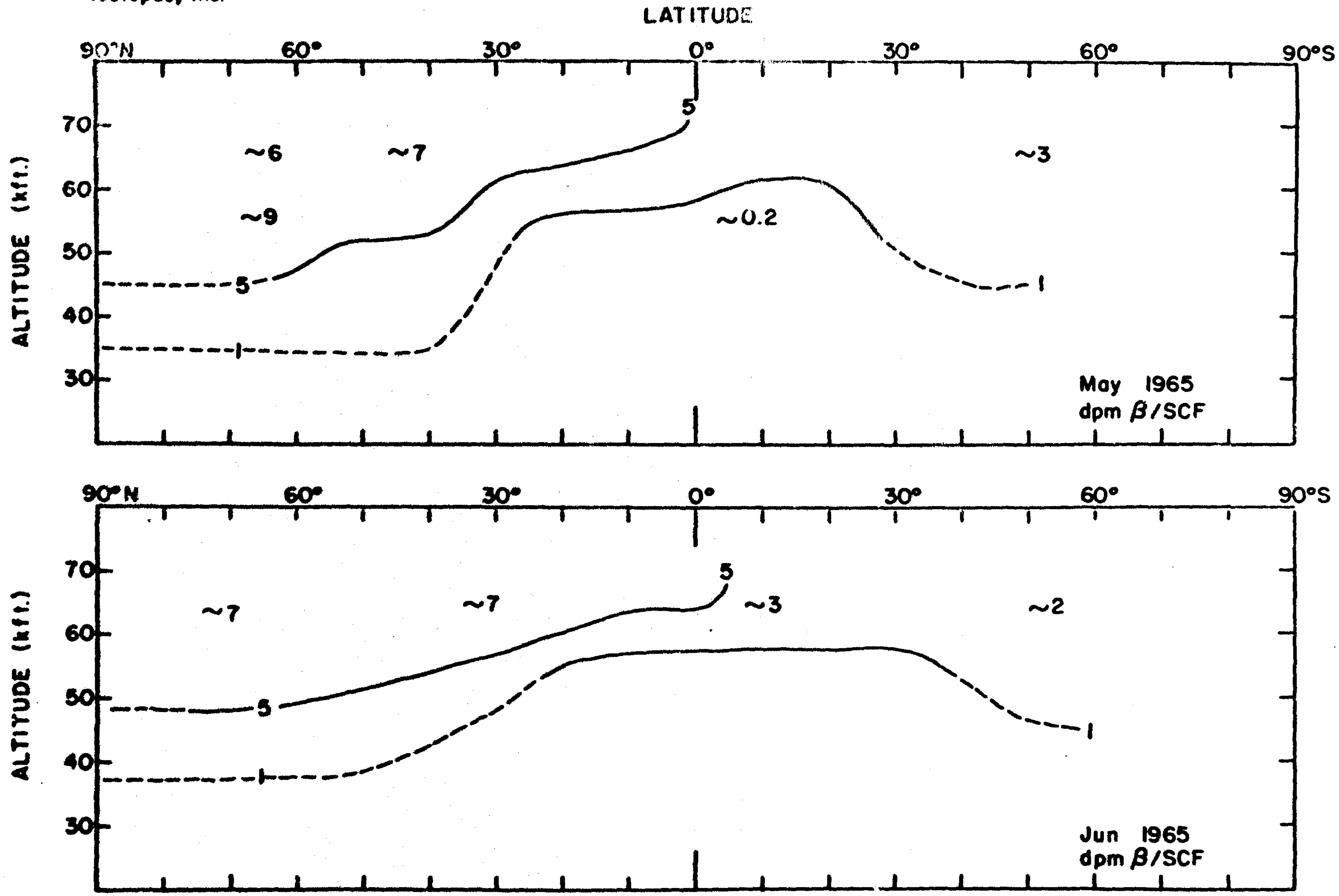


FIGURE 12 · THE MEAN DISTRIBUTION OF TOTAL BETA ACTIVITY DURING MAY AND JUNE 1965

Isotopes, Inc.

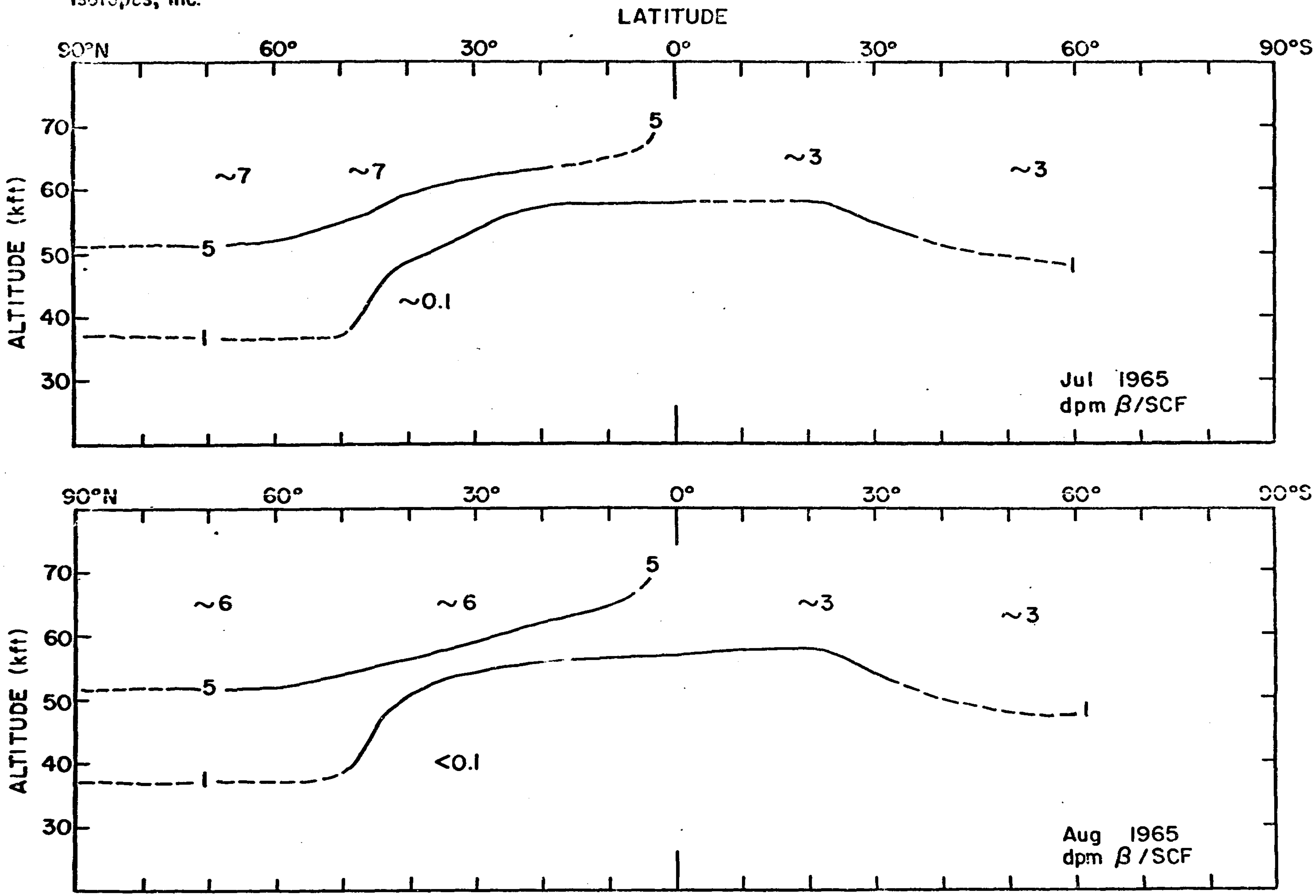


FIGURE 13 · THE MEAN DISTRIBUTION OF TOTAL BETA ACTIVITY DURING JULY AND AUGUST 1965

The Stratospheric Distribution of Strontium-90

The decreases in stratospheric concentrations of strontium-90 which occurred between September 1963 and August 1965 must be attributed mainly to fallout of debris from the stratosphere. Strontium-90 has a relatively long radioactive half life, and concentration decreases as a result of radioactive decay would amount to only about 5 percent during the course of two years. The considerable decreases in the concentrations of strontium-90 in most stratospheric regions during 1963 to 1965 are illustrated by the data in Figures 14 and 15. In these figures are plotted the mean bimonthly concentrations of this nuclide at six locations in the stratosphere: at 50 and at 65 thousand feet at 65° - 70°N, at 55 and at 65 thousand feet at 25°N, and at 55 and at 65 thousand feet at 35° - 40°S.

Vertical profiles of strontium-90 activity at 65° - 70°N, 30° - 35°N, 10°N, and 34° - 40°S during 1963, 1964 and 1965 are plotted in Figures 16, 17, 18 and 19. Data for samples collected at 65°N, 31°N, 9°N and 34°S by the AEC high altitude balloon sampling program[2,3,4] are included in these figures to permit extension of the profiles into the upper stratosphere. At 65° - 70°N the highest strontium-90 concentrations have been found in the layer between 50 and 70 thousand feet fairly consistently since mid-1963. Farther south, at 30° - 35°N, the highest concentrations have been found in the layer between 60 and 85 thousand feet during 1963 to 1965. At 10°N, where balloon measurements have been available only since late 1964, the highest concentrations have been found in the layer between 65 and 90 thousand feet. In the Southern Hemisphere, at 34° - 40°S, the vertical profile of strontium-90 activities displayed a broader maximum, including the layer between 50 and 85 thousand feet.

It would appear from the observed vertical distributions that the bulk of the strontium-90 injected into the atmosphere by the 1961 and 1962 atmospheric

tests of high yield nuclear weapons was injected into the lower 50 thousand feet (30,000 to 80,000 feet) of the polar stratosphere of the Northern Hemisphere, and into the lower 50 thousand feet (50,000 to 100,000 feet) of the tropical stratosphere. Since 1963 both the concentrations of strontium-90 in the stratospheric regions above the level of the maximum and the concentrations in the regions below that level have decreased. It has been suggested that the observed decreases in concentration can be explained only by invoking relatively rapid rates of particle settling in the higher regions of the stratosphere, above 70 thousand feet[5,6].

Some information on the processes of exchange of stratospheric radioactive debris between the Northern and Southern Hemispheres may be obtained from the observed changes in the stratospheric distribution of strontium-90 during 1963 to 1965. In Figure 20 are plotted the bimonthly mean strontium-90 concentrations at 65 thousand feet at 45°N, 0°N and 45°S during these three years. The data for 0°N and 45°S suggest that there was an acceleration in the movement of debris from the Northern into the Southern Hemisphere during May-September, 1963, May-June, 1964, and June-September, 1965: i.e., during the winter and spring seasons of the Southern Hemisphere. It has been suggested[6] that the southward transport of gaseous debris, represented by $C^{14}O_2$, during 1963 at least, was less rapid than that of particulate debris, represented by strontium-90 and manganese-54, and that the difference might be related to supposed differences in the trajectories followed by these two types of debris in moving through the tropical stratosphere.

Combining Stardust strontium-90 data with available data from the USAEC balloon program, mean distributions of strontium-90 have been calculated for a series of four month periods during 1963 to 1965. These are plotted in Figures 21 to 24. During January-April 1963 and May-August 1963 (Figure 21), the highest concentrations of strontium-90 were found in the lower polar stratosphere of the Northern Hemisphere. If balloon data for 34°S can be used

to deduce the distribution of radioactive debris in the upper stratosphere of the Southern Hemisphere, it may be hypothesized that debris from the Starfish Prime high altitude event in the 1962 Dominic series of weapons tests was present in the stratospheric regions above 80 thousand feet in the Southern Hemisphere during January to April 1963. (The use of cadmium-109 for identifying this debris will be discussed in a later section of this chapter.) As a result, there was a layer of minimum strontium-90 concentrations between about 70 and 90 thousand feet in the Southern polar stratosphere during January to April 1963. The regions above this layer contained higher concentrations attributable especially to the Starfish Prime debris, while the regions below it contained higher concentrations attributable mainly to pre-1962 weapons tests series. By May-August 1963 the downward movement of Starfish Prime debris, and especially the southward movement of debris from the 1962 weapons tests had eliminated this layer of low concentrations. By then the highest concentrations of strontium-90 at all latitudes were found in the lower stratosphere.

By September-December 1963 and January-April 1964 (Figure 22), the concentrations of strontium-90 had increased in the lower polar stratosphere of the Southern Hemisphere, and had decreased in the lower polar stratosphere of the Northern Hemisphere. The concentrations had decreased, however, in the upper polar stratosphere of both hemispheres. By May-August 1964 and September-December 1964 (Figure 23), there had been further significant decreases in the concentrations in the lower polar stratosphere of the Northern Hemisphere and in the upper stratosphere of both hemispheres, but only relatively small decreases in concentration had occurred in the lower polar stratosphere of the Southern Hemisphere. By January-April 1965 and May-August 1965 (Figure 24), further significant decreases had occurred in the lower stratosphere of both hemispheres.

The stratospheric burdens represented by the mean distributions shown in Figures 21 to 24 have been calculated, and are shown, together with the

calculated burdens for earlier periods, in Table 2 and in Figure 25. The decrease in the total stratospheric burden during 1963-1965 corresponded to an apparent stratospheric residence half-time of particulate debris of about 10 months. It might have been expected that the apparent residence half-time of strontium-90 would have slowly lengthened with time during 1963 to 1965 as the lower stratosphere was gradually depleted by continued fallout. It has been suggested[6] that its failure to do so may be attributed to the continued downward flux of debris from the upper to the lower stratosphere during 1963-1965 as a result of particle settling.

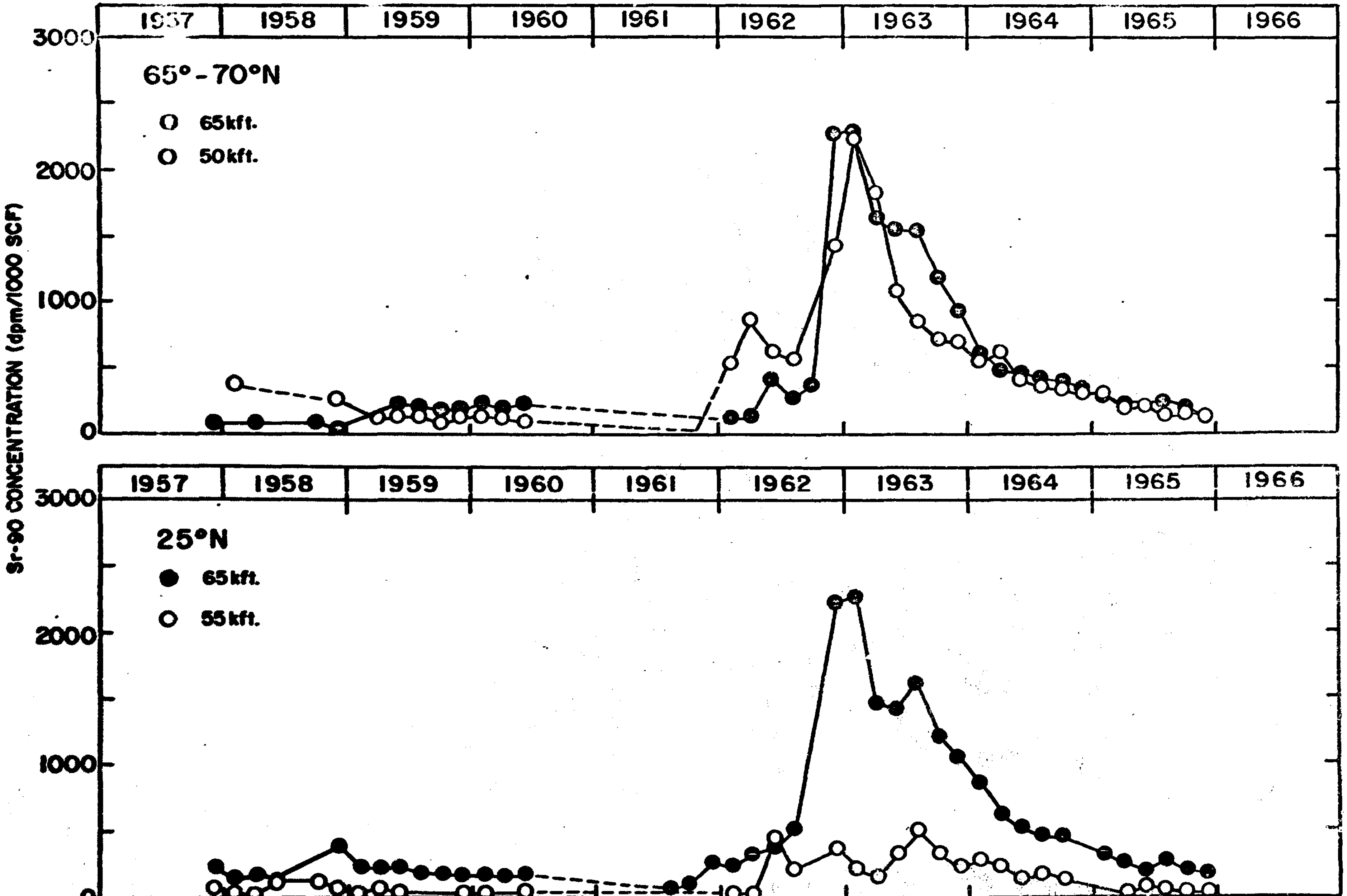


FIGURE 14 · VARIATION WITH TIME OF STRONTIUM-90 CONCENTRATIONS AT 65° - 70°N AND 25°N

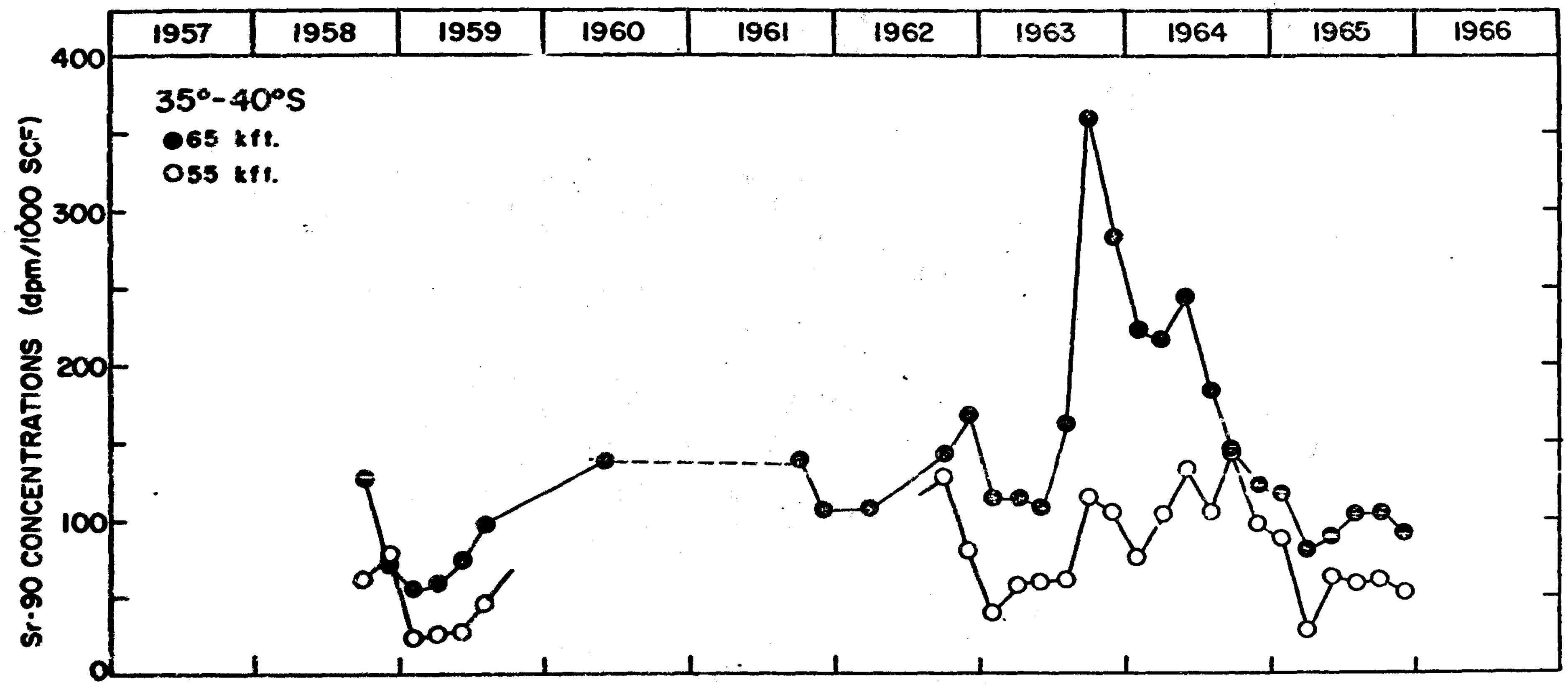


FIGURE 15. VARIATION WITH TIME OF STRONTIUM-90 CONCENTRATIONS AT 35° - 40° S

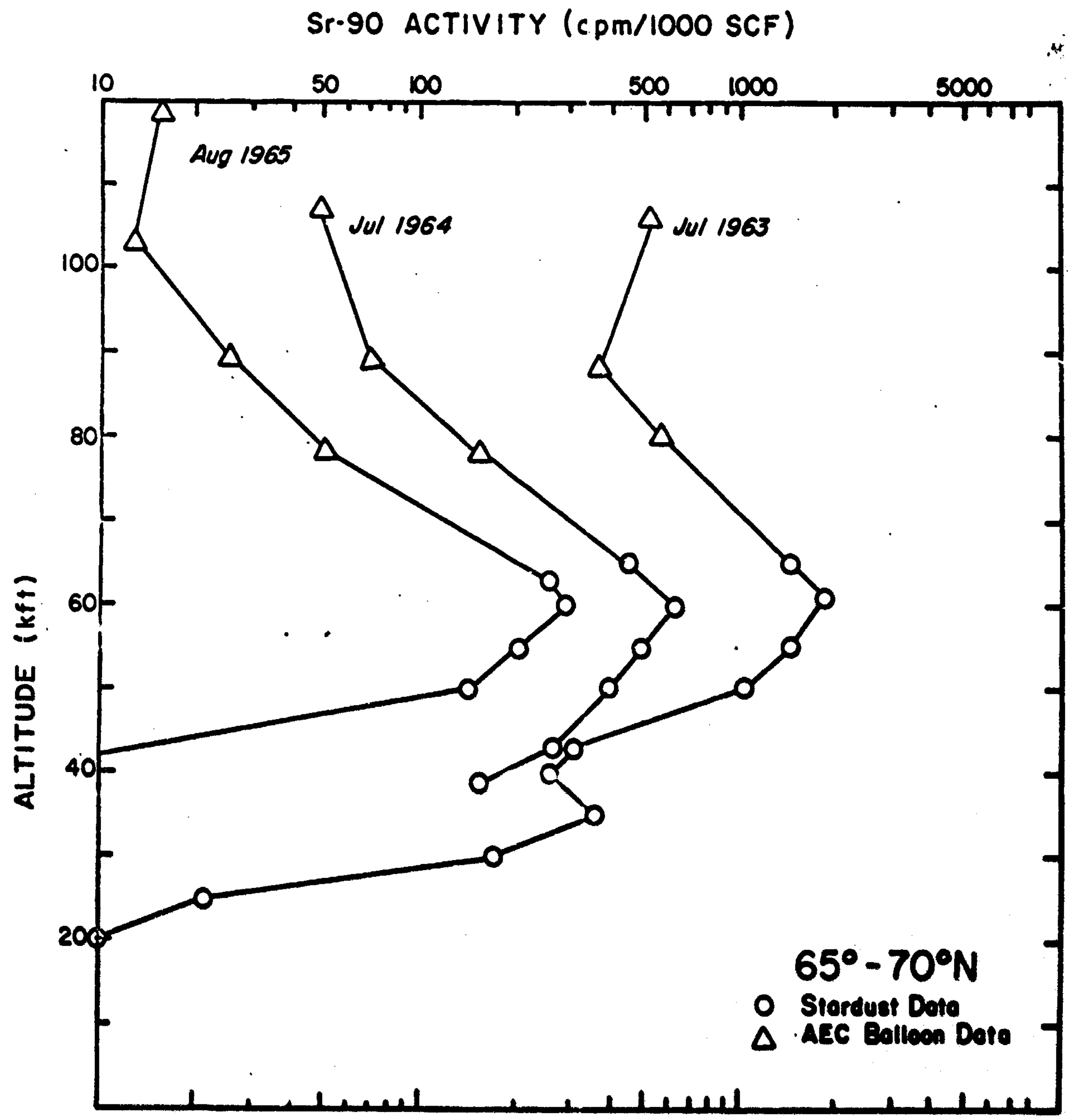


FIGURE 16. VERTICAL PROFILES OF STRONTIUM-90 CONCENTRATIONS AT 65°-70°N

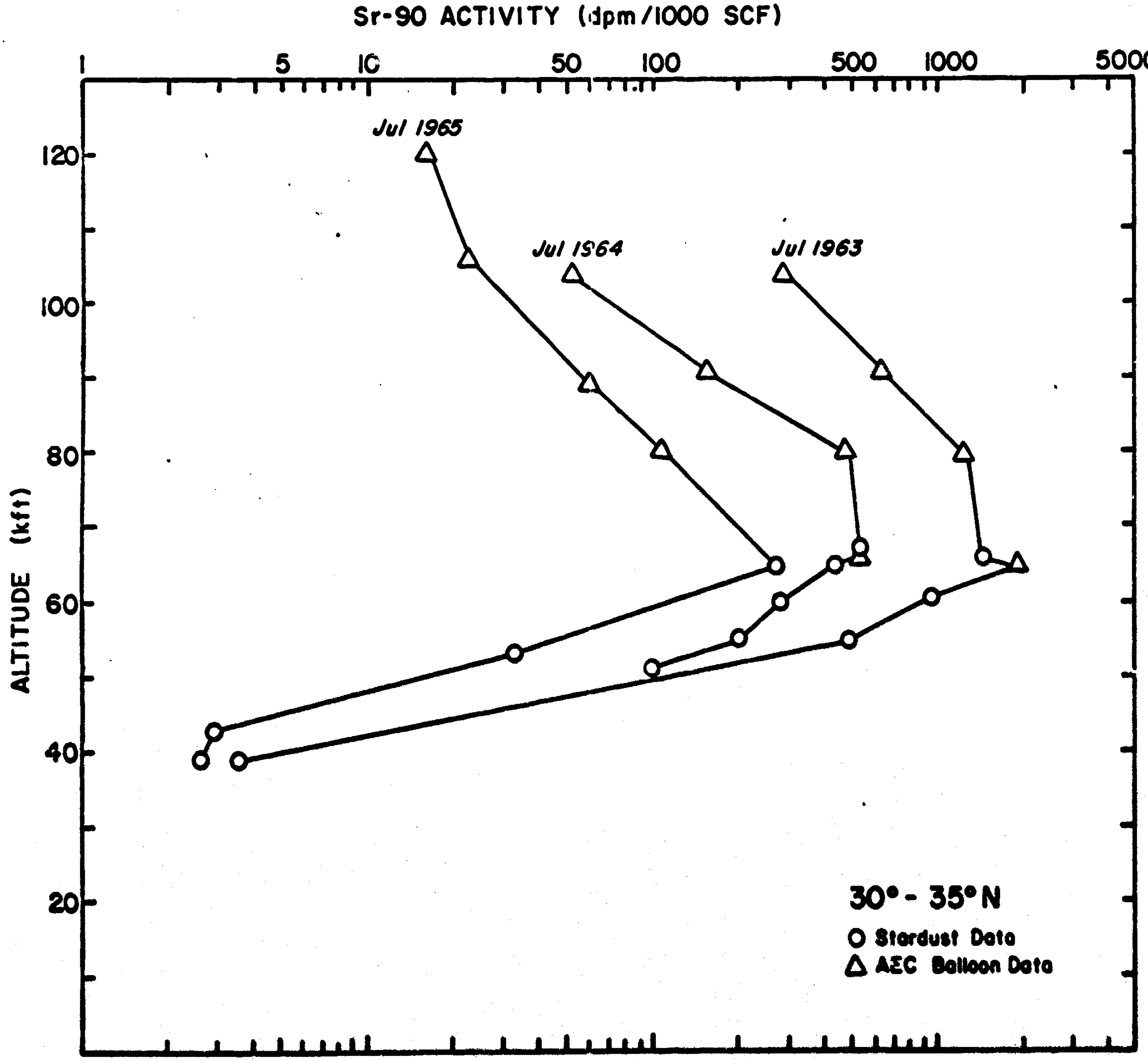


FIGURE 17. VERTICAL PROFILES OF STRONTIUM-90 CONCENTRATIONS AT 30° - 35°N

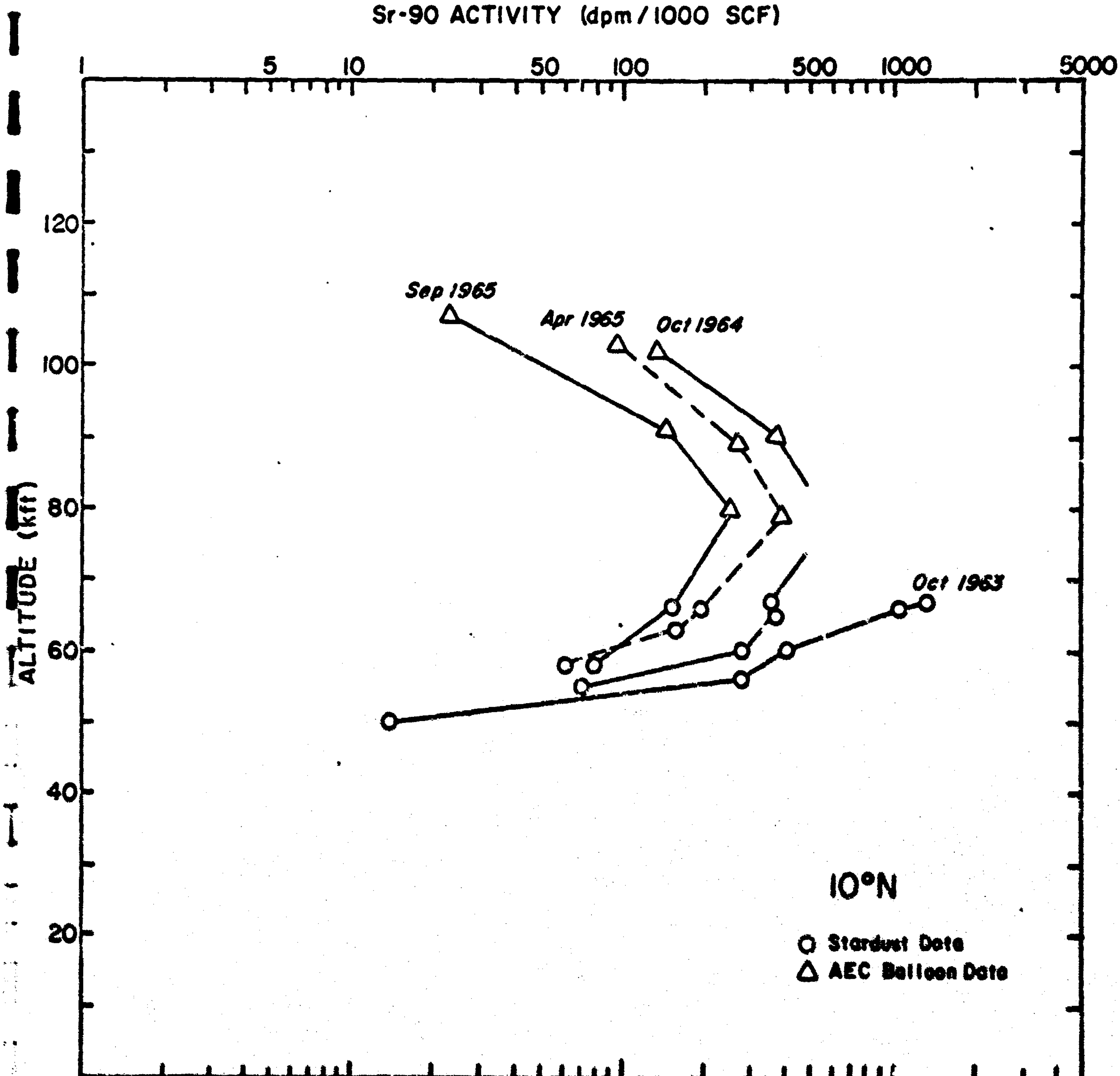


FIGURE 18. VERTICAL PROFILES OF STRONTIUM-90 CONCENTRATIONS AT 10°N

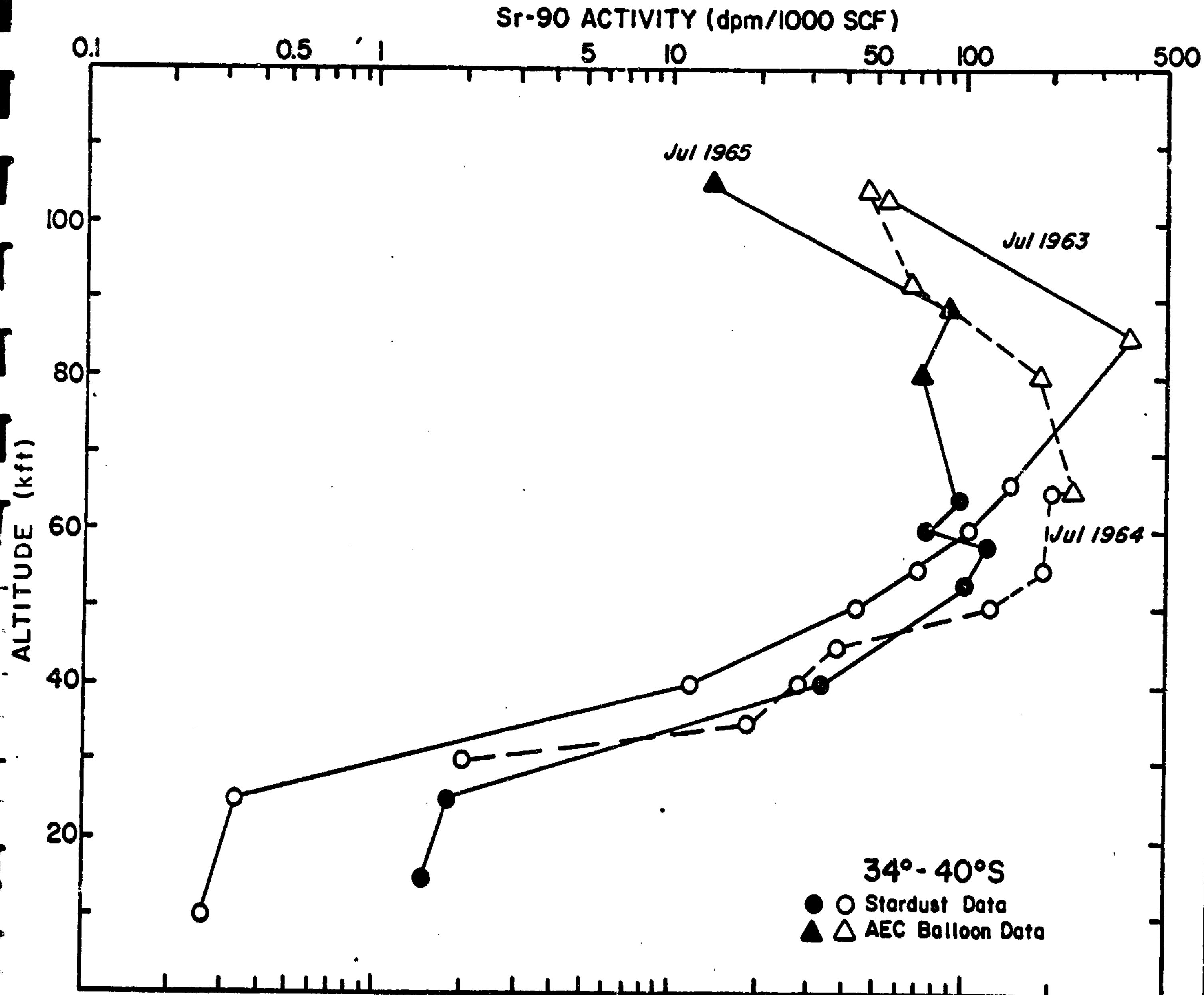


FIGURE 19. VERTICAL PROFILES OF STRONTIUM-90 CONCENTRATIONS AT 34°-40°S

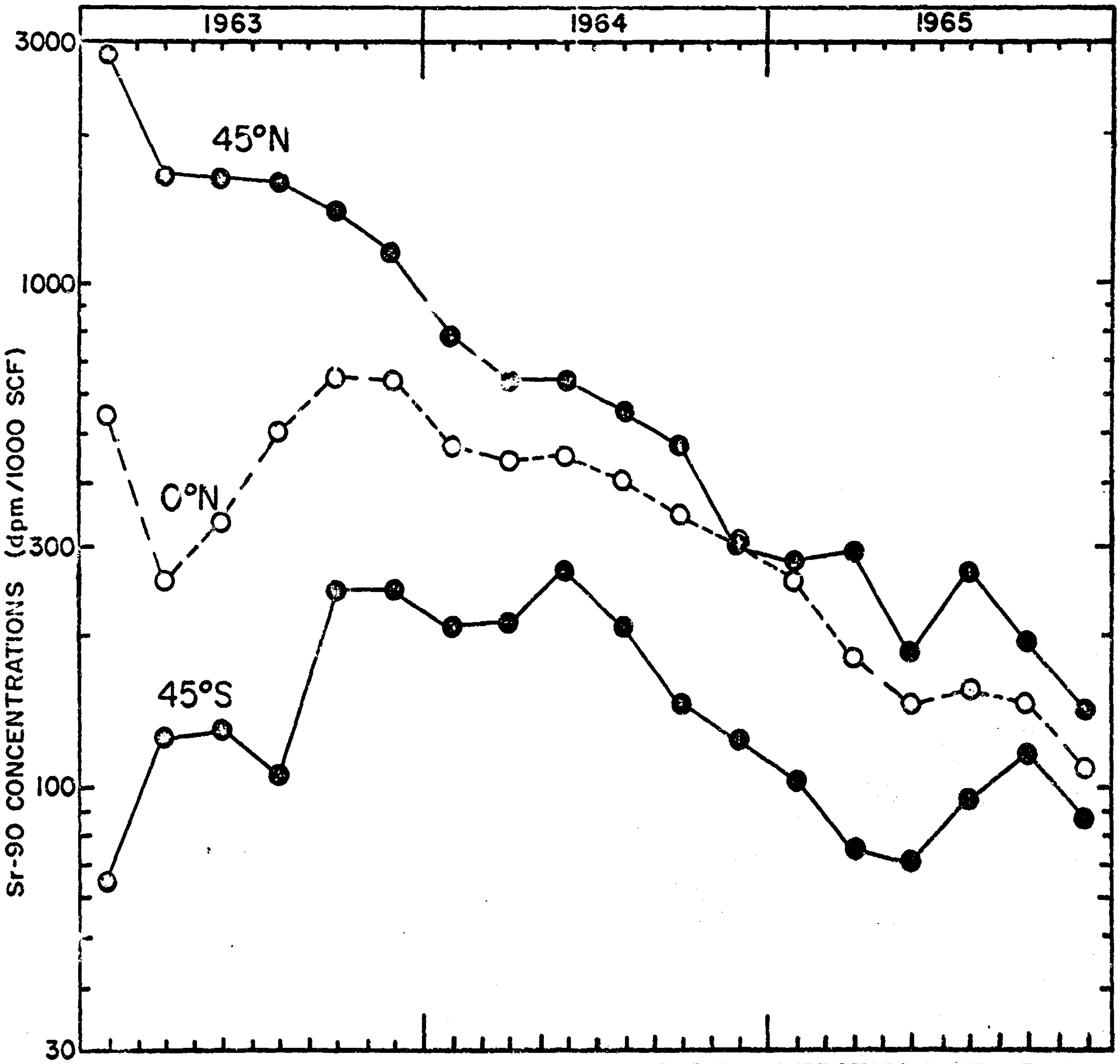


FIGURE 20. TRENDS IN STRONTIUM-90 CONCENTRATIONS AT 65,000 FEET AT 45°N, 0°N AND 45°S DURING 1963 - 1965

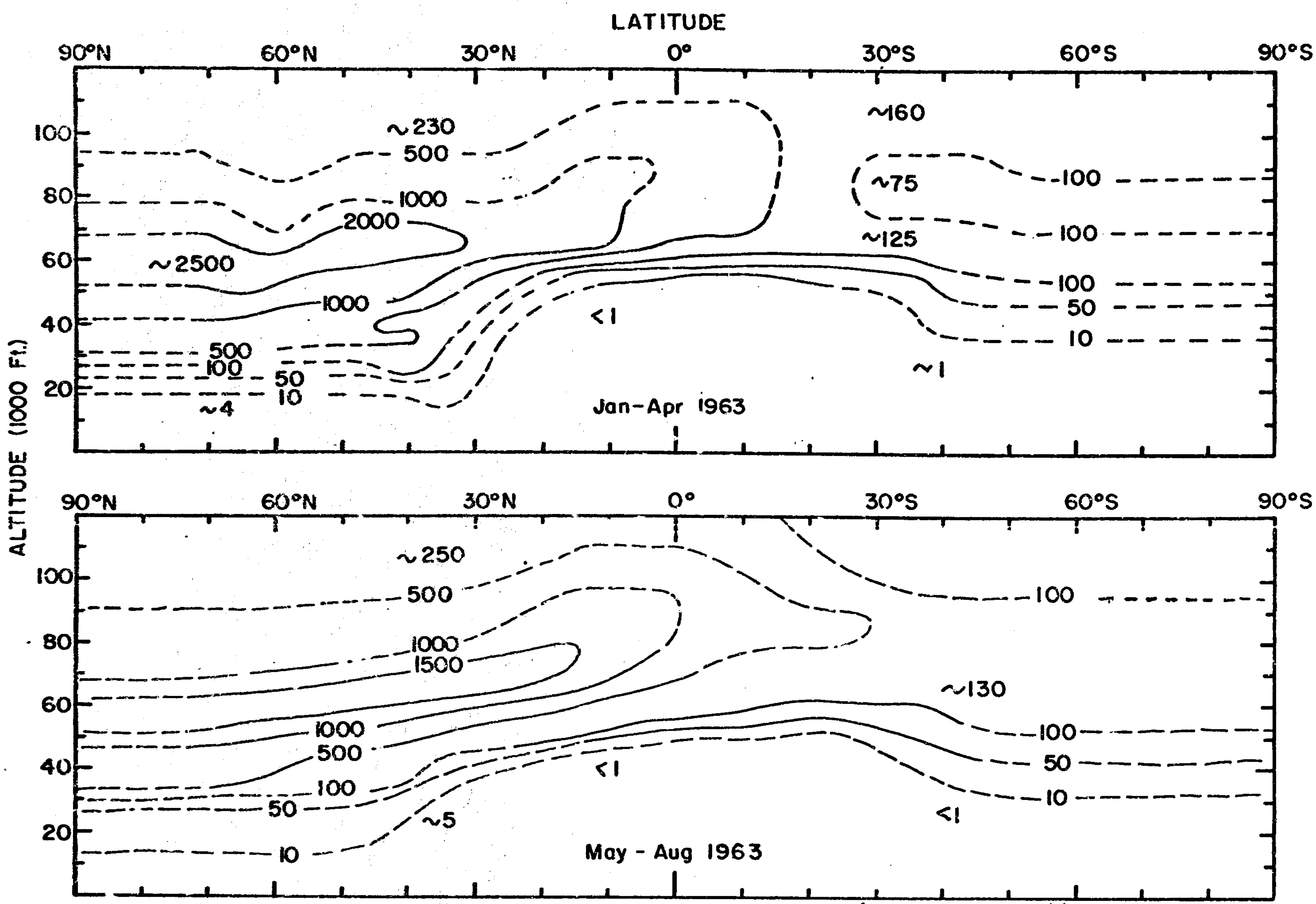


FIGURE 21. THE MEAN DISTRIBUTION OF STRONTIUM-90 (dpm/1000 SCF) DURING JANUARY - APRIL 1963 AND MAY - AUGUST 1963

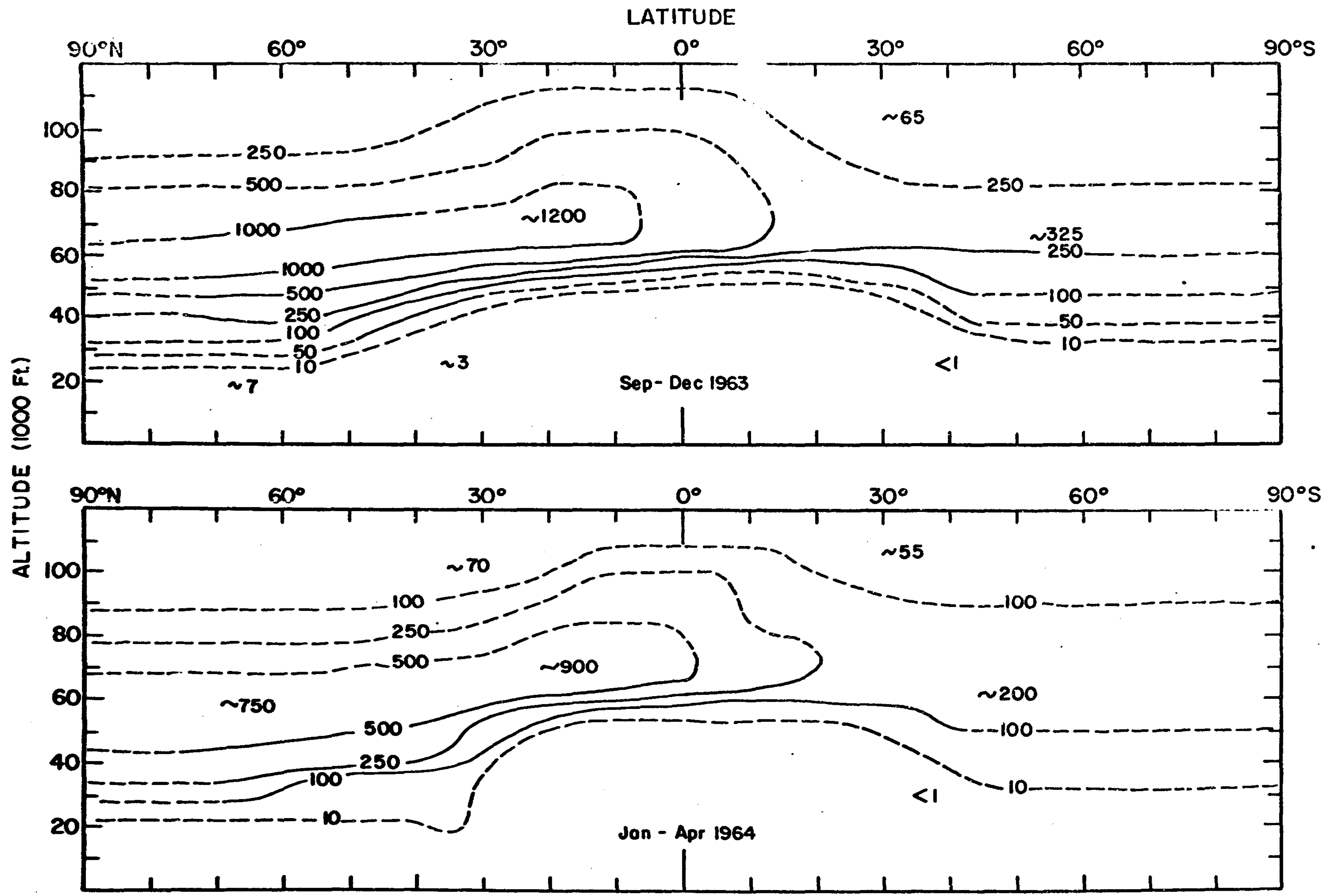


FIGURE 22. THE MEAN DISTRIBUTION OF STRONTIUM-90 (dpm/1000 SCF) DURING SEP. - DEC. 1963 AND JAN. - APR. 1964

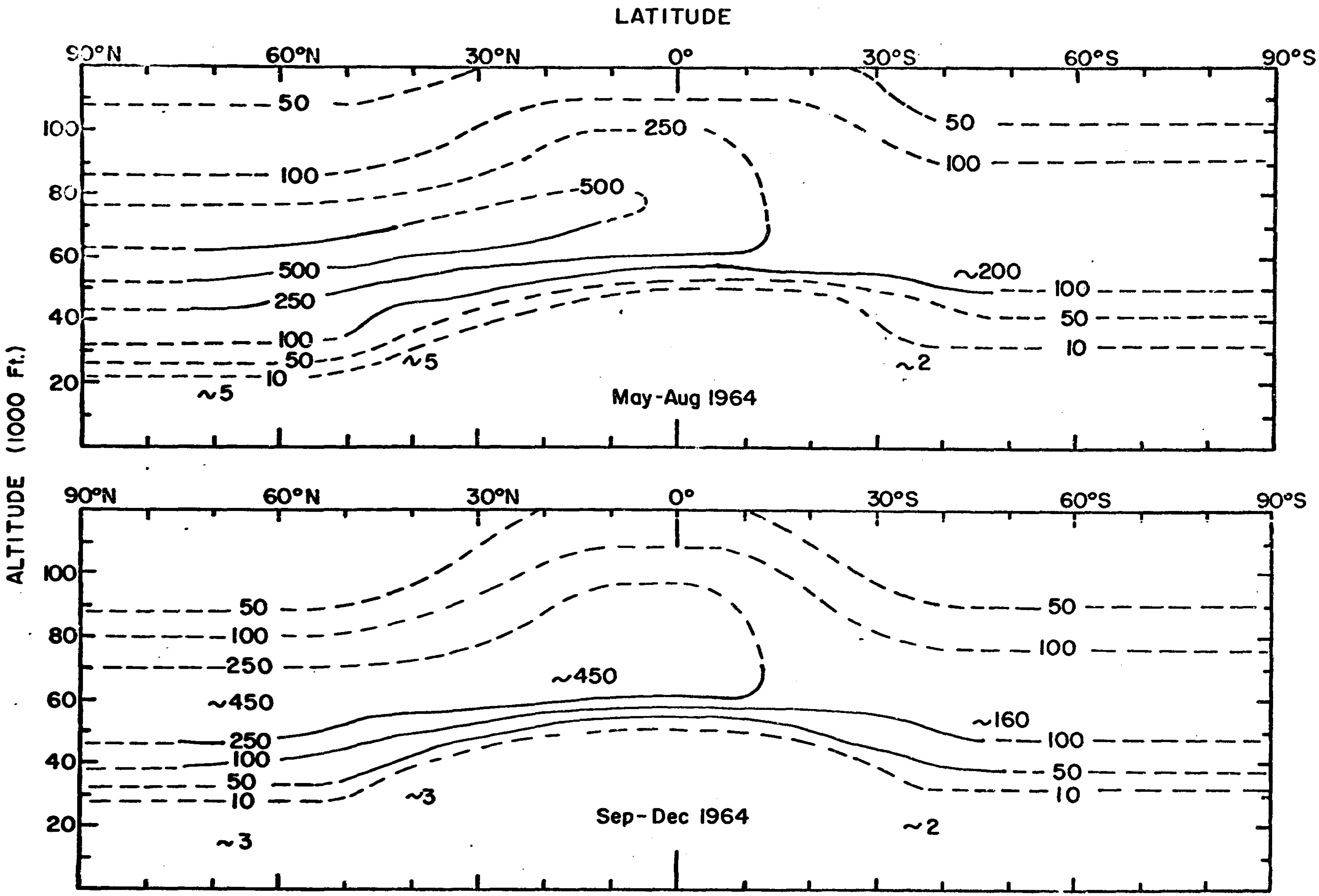


FIGURE 23. THE MEAN DISTRIBUTION OF STRONTIUM-90 (dpm/1000 SCF) DURING MAY-AUGUST 1964 AND SEPTEMBER-DECEMBER 1964

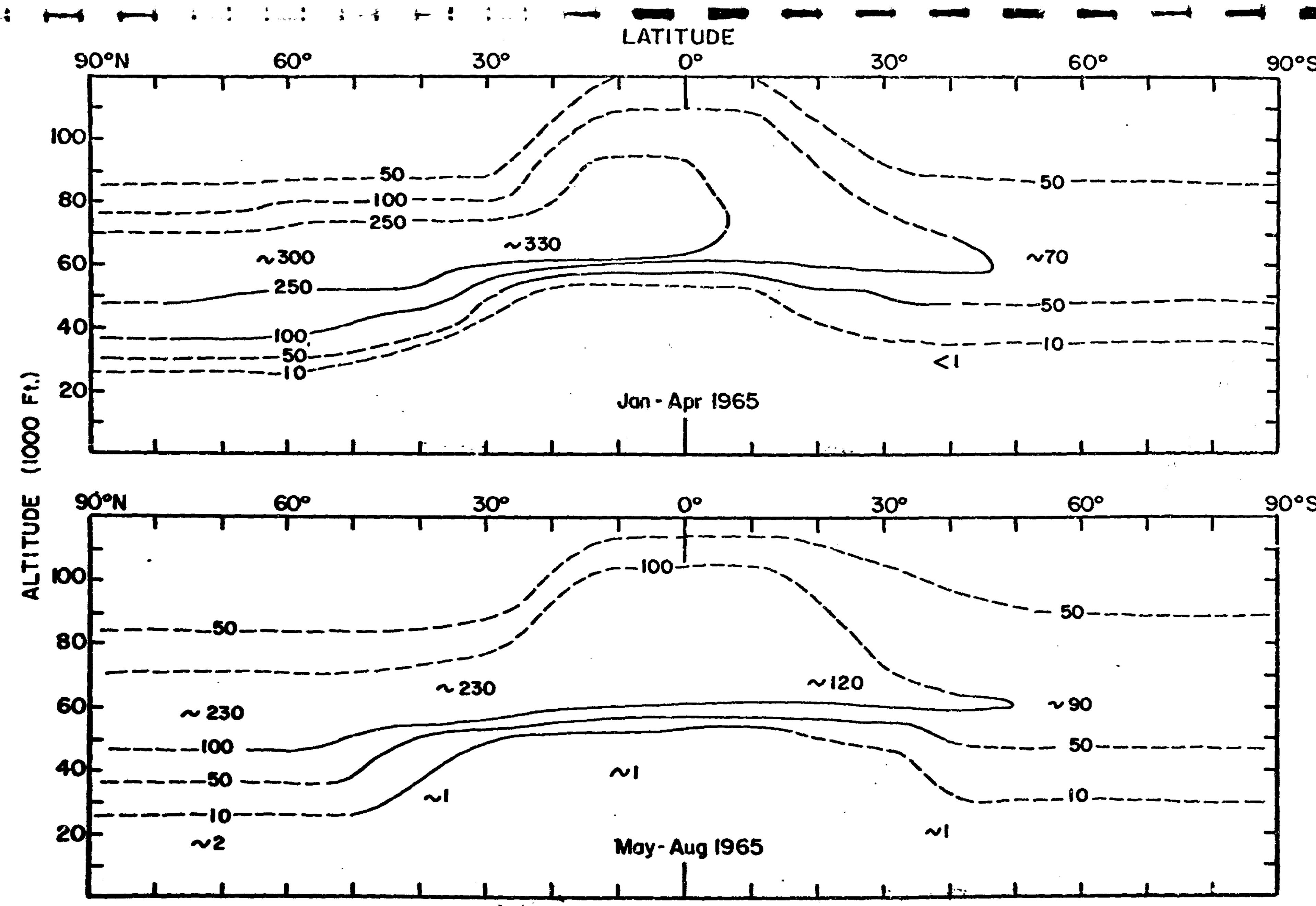


FIGURE 24. THE MEAN DISTRIBUTION OF STRONTIUM-90 (dpm/1000 SCF) DURING JAN - APR 1965 AND MAY - AUG 1965

TABLE 2

THE STRATOSPHERIC BURDENS OF STRONTIUM-90 (MEGACURIES)

Time Interval	Northern Hemisphere	Southern Hemisphere	Total Burden
Jun - Sep 1961	0.37 ± 0.13	~0.5	~0.9
Oct - Dec 1961	~1.6	0.47 ± 0.17	~2.1
Jan - Apr 1962	1.6 ± 0.3	0.4 ± 0.1	2.0 ± 0.3
May - Aug 1962	2.5 ± 0.5	~1.0	~3.5
Sep - Oct 1962	2.2 ± 0.7	1.0 ± 0.3	3.2 ± 0.8
Nov - Dec 1962	5.6 ± 1.9	0.8 ± 0.3	6.4 ± 1.9
Jan - Apr 1963	5.8 ± 1.5	0.7 ± 0.2	6.5 ± 1.5
May - Aug 1963	4.3 ± 1.1	0.8 ± 0.3	5.1 ± 1.2
Sep - Dec 1963	2.8 ± 0.6	1.0 ± 0.3	3.8 ± 0.7
Jan - Apr 1964	2.3 ± 0.4	0.7 ± 0.3	3.0 ± 0.5
May - Aug 1964	1.5 ± 0.4	0.6 ± 0.3	2.1 ± 0.5
Sep - Dec 1964	1.1 ± 0.4	0.6 ± 0.3	1.7 ± 0.5
Jan - Apr 1965	0.9 ± 0.3	0.4 ± 0.2	1.3 ± 0.3
May - Aug 1965	0.6 ± 0.3	0.3 ± 0.2	0.9 ± 0.4

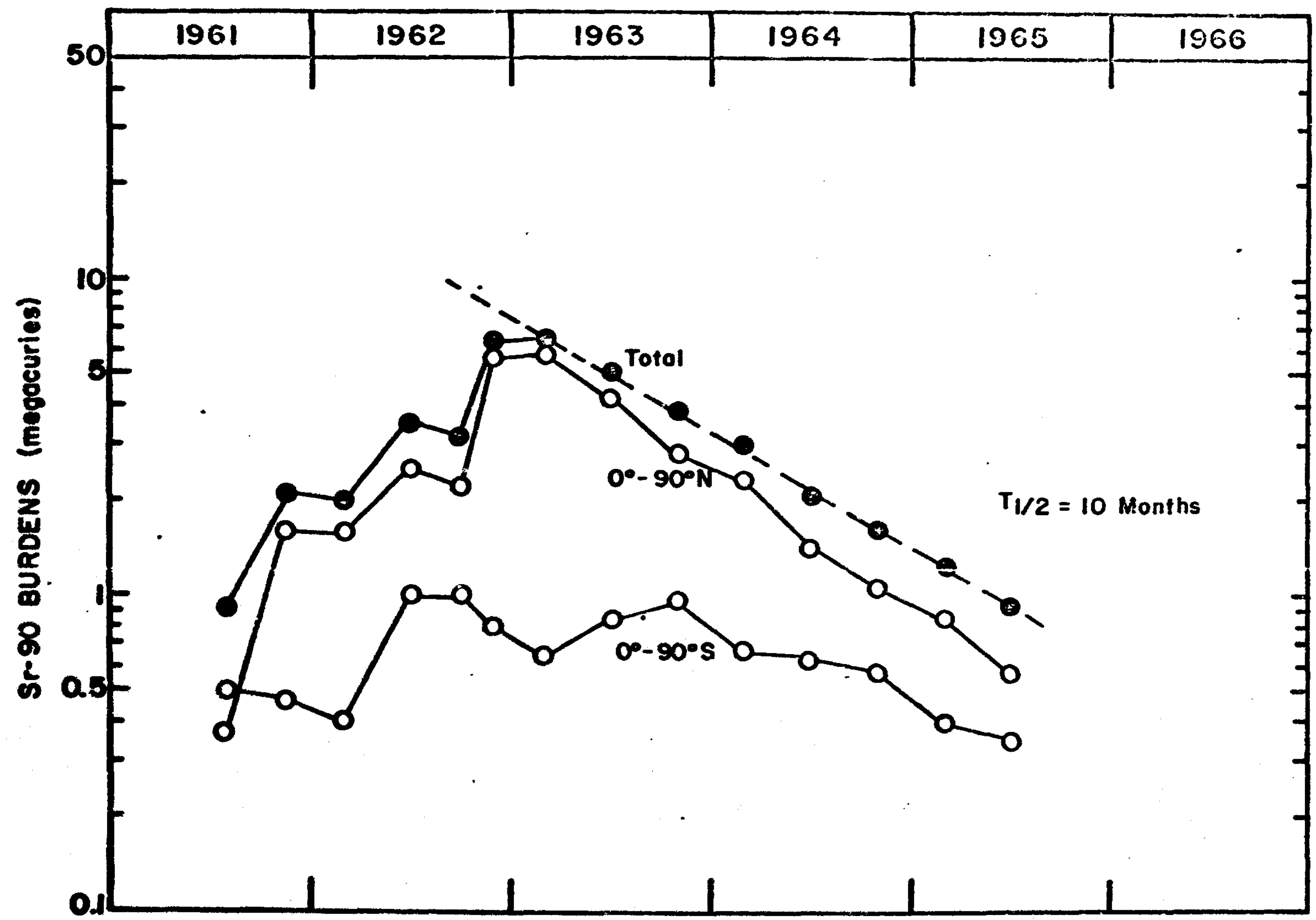


FIGURE 25. STRATOSPHERIC BURDENS OF STRONTIUM-90

The Stratospheric Distribution of Cesium-137

Cesium-137, like strontium-90, is a long-lived fission product which has been injected into the stratosphere by past nuclear weapon tests. Assuming that the relative production rates of cesium-137 and strontium-90 have been similar in all past nuclear weapon tests, we would expect that the Cs^{137}/Sr^{90} activity ratio would be approximately constant in all Stardust filter samples. The available data tend to corroborate this expectation.

Cesium-137 has been measured in 174 Stardust filter samples: in 23 collected during June, 1961 to April, 1962, in 57 collected during May to December, 1962, in 50 collected during 1963, and in 44 collected during 1964. The frequency distribution of measured Cs^{137}/Sr^{90} ratios is shown in Figure 26. The mean value of the ratio for all samples is 1.56 ± 0.24, and the mean value for each of the four intervals specified above falls within one standard deviation of this overall mean. Thus a fairly accurate estimate of the cesium-137 burden of the stratosphere may be obtained by multiplying the strontium-90 burden, given in the preceding section of this chapter, by 1.56.

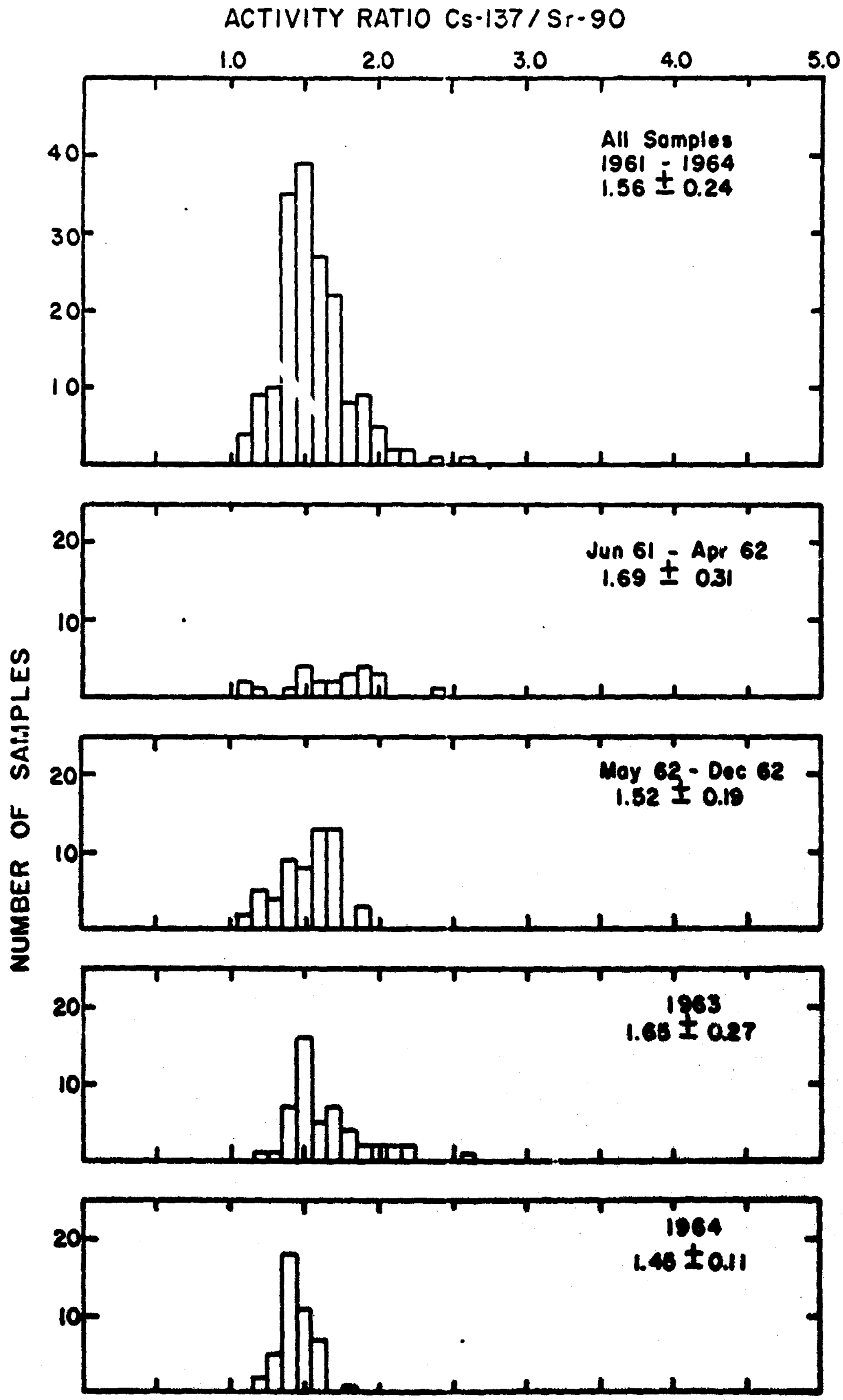


FIGURE 26. FREQUENCY OF Cs-137/Sr-90 RATIOS IN STARDUST SAMPLES

The Stratospheric Distribution of Plutonium-239

Plutonium-239 may have been an original constituent of many of the nuclear devices tested during past years. Additional plutonium-239 may have been produced during the testing of many high yield devices if they contained uranium-238. In such weapons uranium-239 could have been produced by an (n, γ) reaction, and the 23.5 minute uranium-239 would have decayed to 2.35 day neptunium-239 by β^--emission. The latter would, in turn, have decayed by β^--emission to plutonium-239. Both the original weapon materials and the products of neutron activation could include plutonium-240 as well as plutonium-239.

A number of Stardust samples have been measured for plutonium-239. Actually the analytical results represent the combined activities of plutonium-239 and plutonium-240, for the α-spectrometer measurements do not distinguish between radiation from these two nuclides. Our references to "plutonium-239" in this chapter and the next thus actually refer to plutonium-239 plus plutonium-240.

The frequency distributions of measured Pu^{239}/Sr^{90} ratios in samples collected during 1961 to 1965 are shown in Figure 27, broken down into three groups according to latitude of collection. A total of 158 samples, with a mean ratio of 0.017, were collected in the region between 90°N and 30°N. A total of 114 samples, with a mean ratio of 0.015, were collected in the region between 30°N and 35°S. A total of 37 samples, with a mean ratio of 0.014, were collected in the region between 35°S and 60°S. Much of the observed range in values of the Pu^{239}/Sr^{90} ratio doubtless results from real differences in the production rates of these nuclides by individual events in past series of nuclear weapons tests. Nevertheless, there has been little variation of any significance in the Pu^{239}/Sr^{90} ratios observed in Stardust samples, either as a function of latitude or of time of collection, for all Stardust samples contain a mixture of debris from many sources. One exception to this generalization, however, is the low ratio found in samples collected during May-December 1962,

and especially in the tropical stratosphere. The frequency distribution of Pu^{239}/Sr^{90} ratios measured in these samples, in which the mean value of the ratio was 0.009 ± 0.004, is compared in Figure 28 with the frequency distribution of this ratio in all 309 samples measured during 1961-1965, and having a mean value of this ratio of 0.016 ± 0.005.

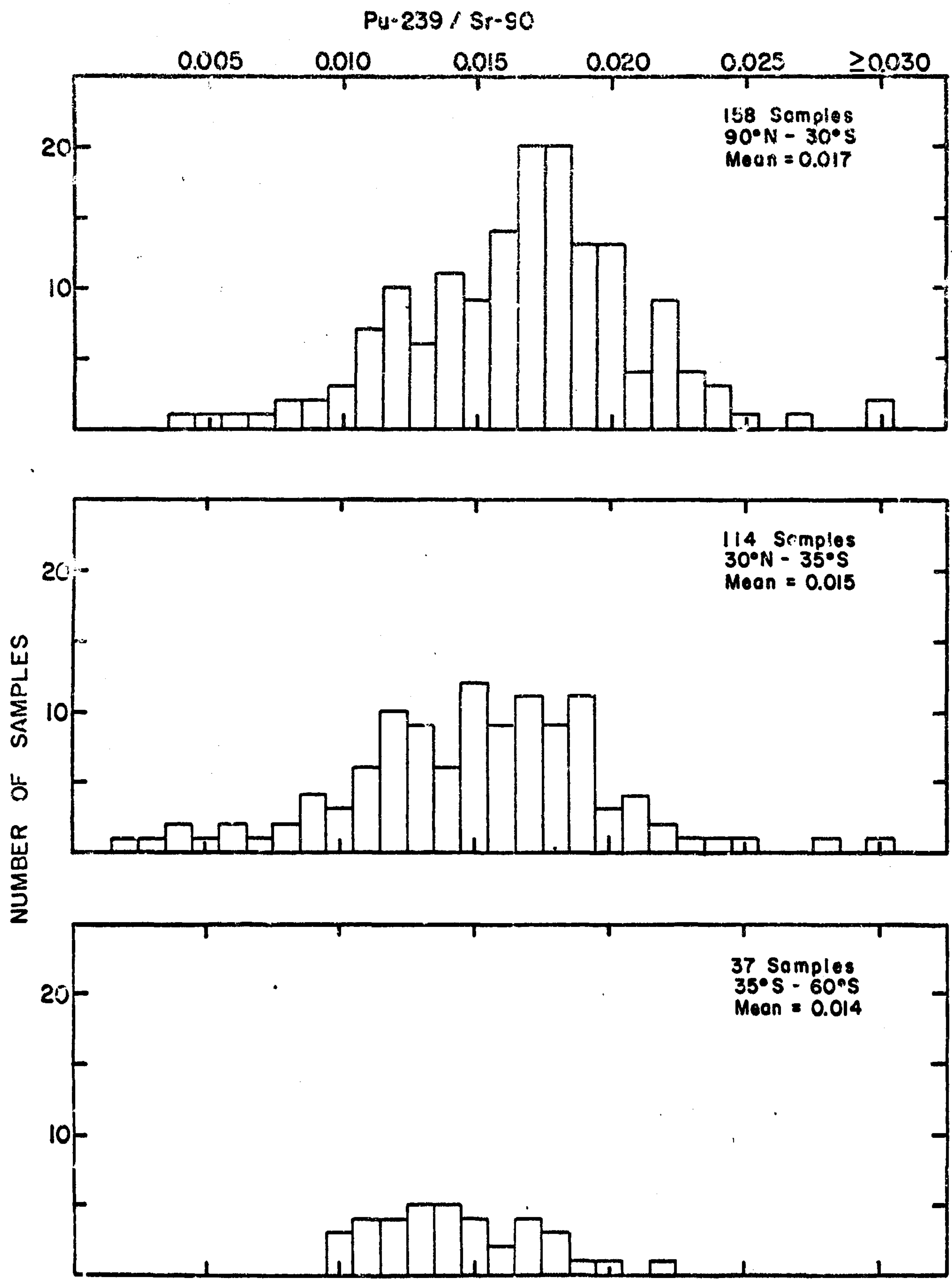


FIGURE 27. FREQUENCY OF Pu-239/Sr-90 RATIOS IN STARDUST SAMPLES ACCORDING TO LATITUDE OF COLLECTION

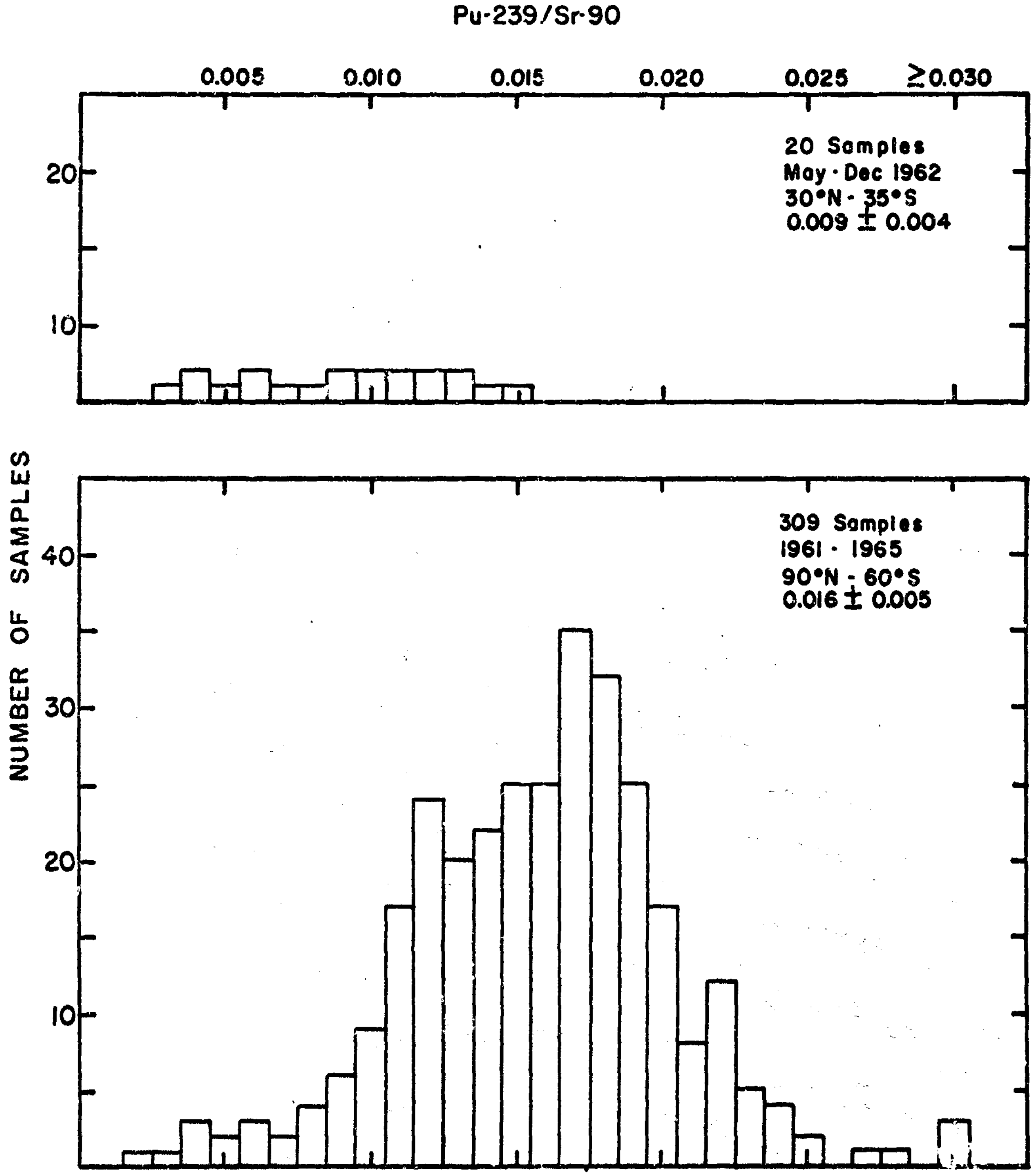


FIGURE 28. FREQUENCY OF Pu-239/Sr-90 RATIOS IN ALL STARDUST SAMPLES AND IN SAMPLES FROM LOW LATITUDES DURING MID- TO LATE 1962

The Stratospheric Distribution of Manganese-54, Iron-55 and Antimony-124

In the Ninth Quarterly Report on Project Stardust, DASA-1309, the stratospheric distributions of several products of neutron activation which are present in stratospheric radioactive debris were discussed. These radionuclides, including manganese-54, iron-55 and antimony-124, were produced in unusually large quantities by certain events during the 1961 and 1962 series of atmospheric tests of nuclear weapons. Apparently the most important sources were the very high yield events during the U.S.S.R. test series.

During 1963 to 1965, the changes which have taken place in the stratospheric distributions of manganese-54 and of the other activation products have been similar to the changes which have taken place in the distribution of strontium-90: concentrations of the activation products, corrected for radioactive decay to 31 December 1962, have decreased in the stratosphere of the Northern Hemisphere, initially increased but subsequently decreased in the stratosphere of the Southern Hemisphere, and decreased continuously in the stratosphere as a whole.

Vertical profiles of manganese-54 concentrations at about 65°N, 35°N and 35°S during August 1964 have been calculated using data from the USAEC balloon program[3] as well as from Project Stardust. They are shown in Figure 29. At 65°N and 35°S the highest concentrations of this nuclide were found in the lower stratosphere in the layer between 45 or 50 thousand feet and 65 thousand feet. At 35°N the layer between 60 and 90 thousand feet contained the highest concentrations. Samples collected above 100 thousand feet at all three latitudes contained concentrations below the limits of detection.

Using a value of 1000 dpm Mn^{54}/1000 SCF (corrected for decay to 31 December 1962) to demarcate the layer of maximum concentration in the

Northern Hemisphere, we find that this layer is approximately 65 millibars in thickness at 65°N, lying between 49 thousand feet (122 mb.) and 65 thousand feet (57 mb.), and approximately 68 millibars in thickness at 35°N, lying between 57 and 93 thousand feet (i.e., 83 and 15 mb.). This similarity in thickness (while to some extent it may be fortuitous) is consistent with the concept that meridional movement occurs readily within the polar stratosphere along surfaces which slope upward toward the equator, but that vertical movement of debris, at least during the summer season, is slow in rate. One would thus expect, as is confirmed by Figure 29, that at all latitudes within the polar stratosphere, between about 30° and 90° latitude, approximately equal concentrations would be found within the layer of maximum concentration, but that this layer would be found to be thicker and to occur at higher altitudes as one moved from the pole toward the equator.

The mean distribution of manganese-54 in the stratosphere has been calculated for each of eight periods of four month duration during 1963 to 1965, and these are shown in Figures 30 to 33. During January-April, 1963 and May-August 1963 (Figure 30), almost all of the manganese-54 produced by the 1961 and 1962 series of weapons tests was still present in the stratosphere of the Northern Hemisphere, though southward movement of this debris had already caused significant changes in its stratospheric distribution by mid-1963. By September-December 1963 and January-April 1964 (Figure 31), continued southward movement of debris had produced increased concentrations of manganese-54 in the lower southern polar stratosphere, and, combined with fallout of debris, had produced large decreases in concentrations in the lower northern polar stratosphere. These trends continued during May-August 1964 and September-December 1964 (Figure 32), with further decreases in concentration occurring in the lower northern polar stratosphere. In the lower southern polar stratosphere, however, fallout of manganese-54 more or less balanced its influx from the Northern Hemisphere during 1964, and there was relatively

little change in the total amount of manganese-54 in that region during the year. By January-April 1965 and May-August 1965 (Figure 33), however, fallout became the predominant factor in the Southern as well as the Northern Hemisphere, and both hemispheres were characterized by decreasing concentrations.

During 1963 to 1965 decreases occurred in the manganese-54 concentrations in the upper stratosphere as well as in the lower stratosphere; i.e., in the regions above the layer of maximum concentration, as well as in that layer and in the regions below it. Again, as with the observed decreases in strontium-90 concentrations, these decreases may be taken as evidence for the importance of particle settling as a mechanism for the transfer of radioactive debris from the upper into the lower stratosphere[5,6].

Stratospheric burdens of manganese-54 have been calculated from each of the mean distributions shown in Figures 30 to 33. The results are summarized in Table 3 and are plotted in Figure 34. It appears that manganese-54, like strontium-90, displayed a residence half-time of about 10 months during 1963 to 1965.

A number of samples were measured for iron-55 as well as for manganese-54. Since the mechanisms of production of these two nuclides in nuclear weapon debris are probably closely related ($Fe^{54}(n, \gamma)Fe^{55}$, $Fe^{54}(n, p)Mn^{54}$, $Fe^{56}(n, 2n)Fe^{55}$, $Mn^{55}(n, 2n)Mn^{54}$), it is not too surprising that a fairly constant ratio of iron-55 to manganese-54 activities (both corrected for decay to 31 December 1962) was found in Stardust samples collected during 1963 to 1965. The mean value of the Fe^{55}/Mn^{54} ratio found in 474 samples was 2.4 ± 0.4. The mean value found in the 277 samples collected between 90°N and 30°N was 2.3 ± 0.4. In the 111 samples collected between 30°N and 10°S the ratio was 2.4 ± 0.4, and in the 86 samples collected between 15°S and 60°S the ratio was 2.5 ± 0.5.

Antimony-124, another product of neutron activation ($Sb^{123}(n, \gamma)Sb^{124}$), was produced in large quantities by one or more events in the weapons tests performed

by the USSR in December, 1962. The mean distributions of antimony-124 concentrations, corrected for decay to 31 December 1962, during January-April 1963, May-August 1963 and September-December 1963 are shown in Figure 35. By early 1964 the radioactive decay of this nuclide, which has a half life of 60.2 days, had reduced concentrations in most stratospheric regions to values too low to be easily measured accurately, and measurements of it in Stardust samples were soon abandoned. During early 1963 the antimony-124 rapidly achieved a rather uniform distribution with latitude within the northern polar stratosphere, 90°N-30°N, and some entered the tropical stratosphere. During the remainder of the year increasing quantities entered the tropical stratosphere, but little had reached the Southern Hemisphere by the end of 1963.

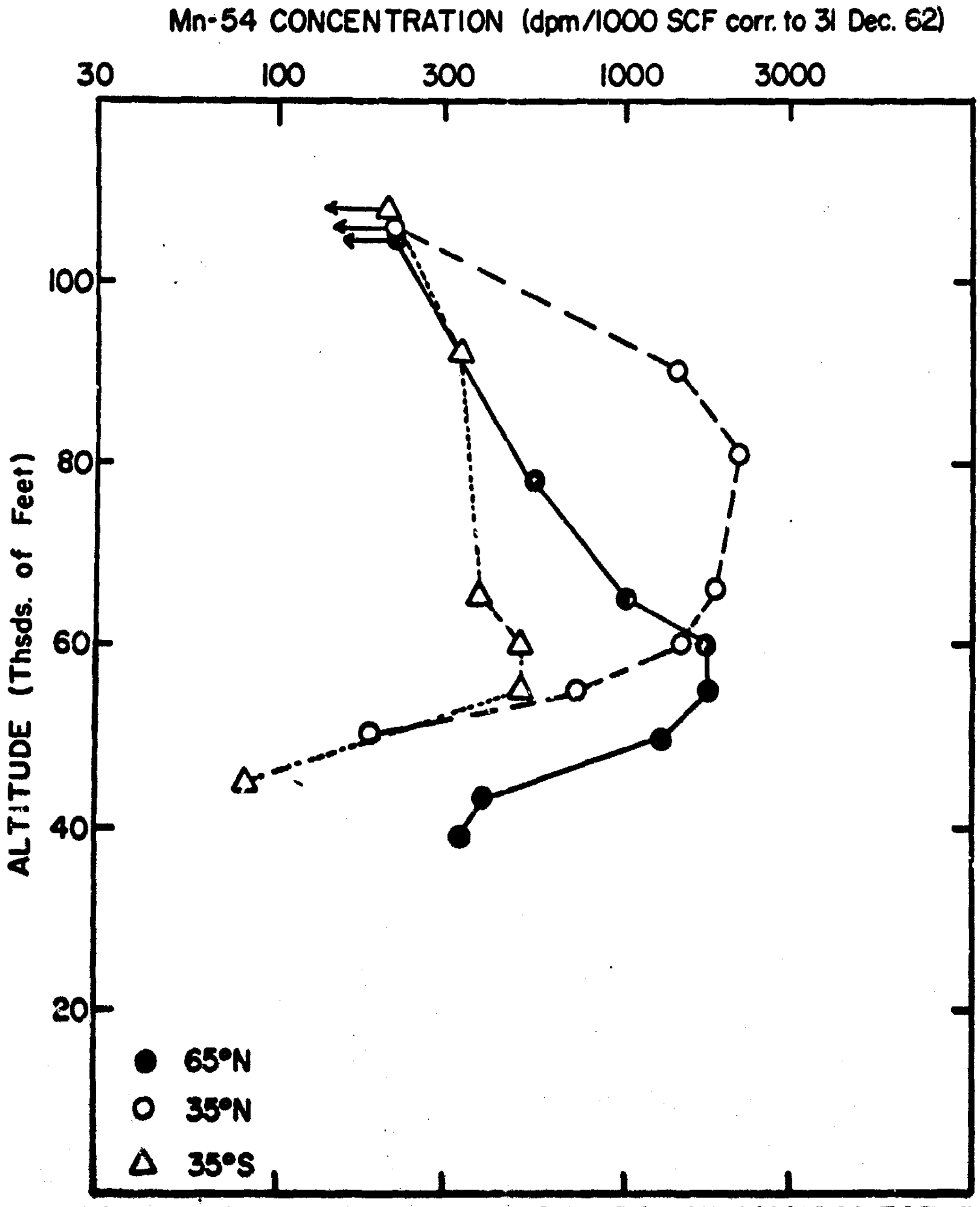


FIGURE 29. VERTICAL PROFILES OF MANGANESE-54 DURING AUGUST 1964

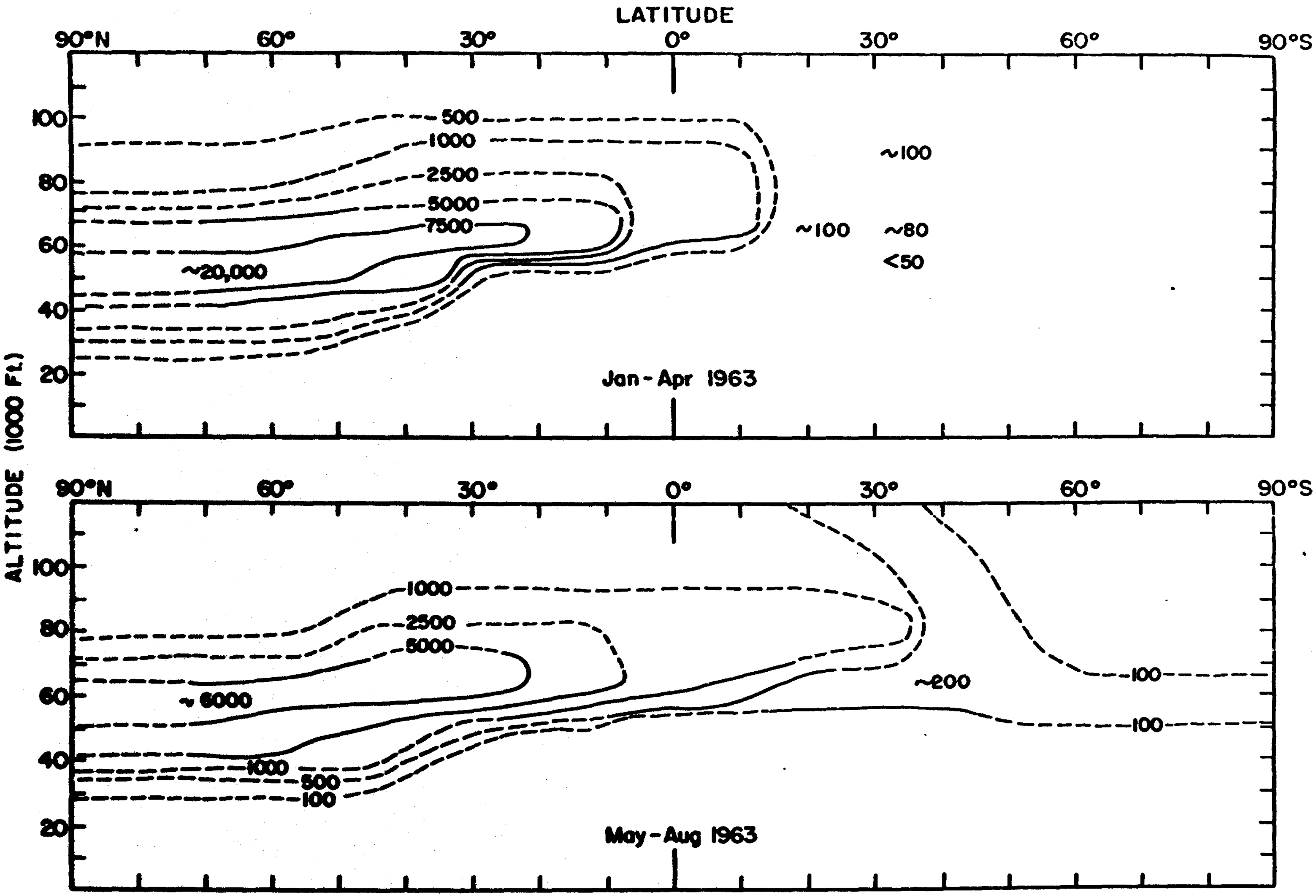


FIGURE 30. THE MEAN DISTRIBUTION OF MANGANESE 54 (dpm/1000 SCF, corr. for decay to 31 Dec 1962) DURING JAN-APR 1963 AND MAY-AUG 1963

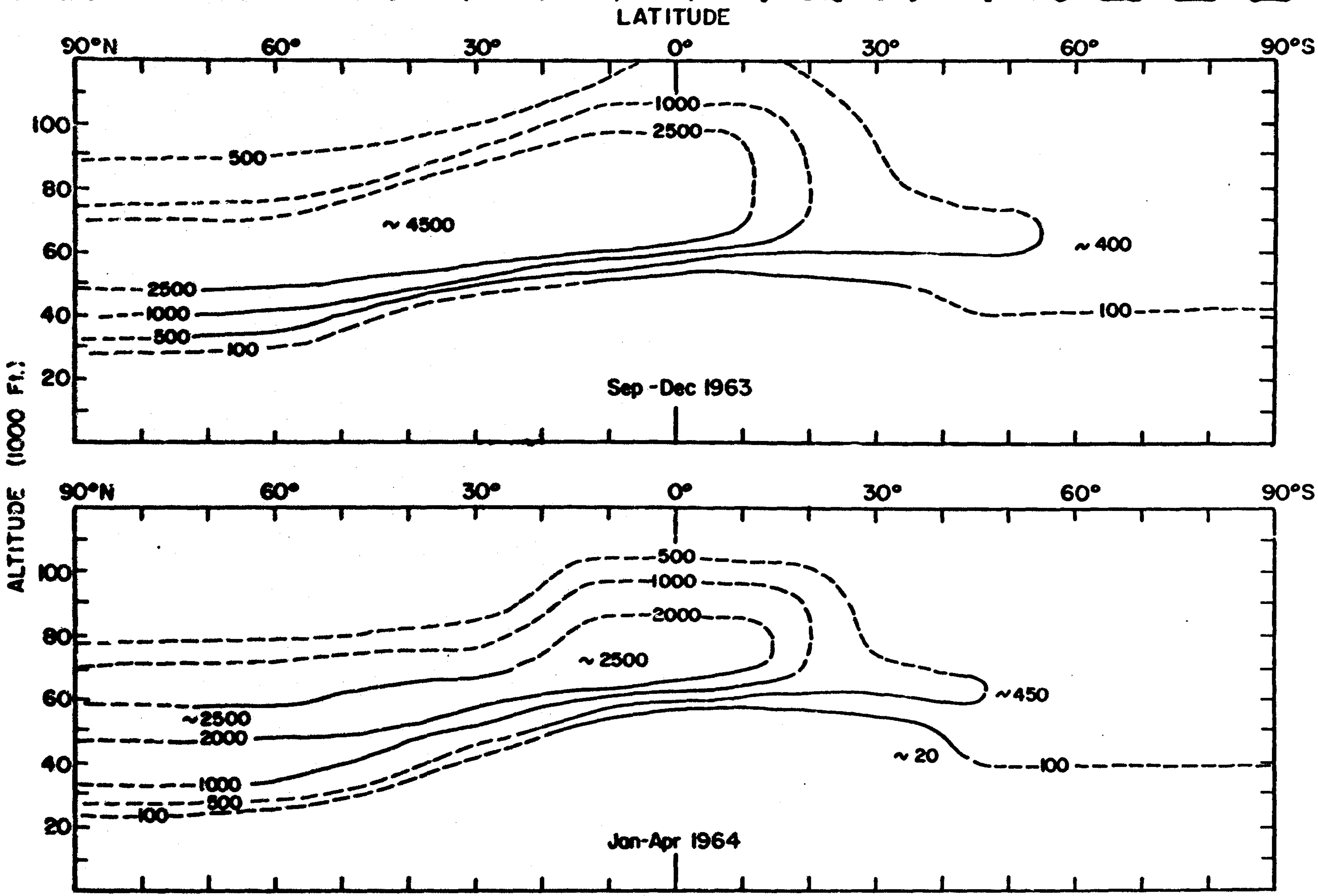


FIGURE 31. THE MEAN DISTRIBUTION OF MANGANESE-54 (dpm/1000 SCF corr. for decay to 31 Dec 1962) DURING SEP-DEC 1963 AND JAN-APR 1964

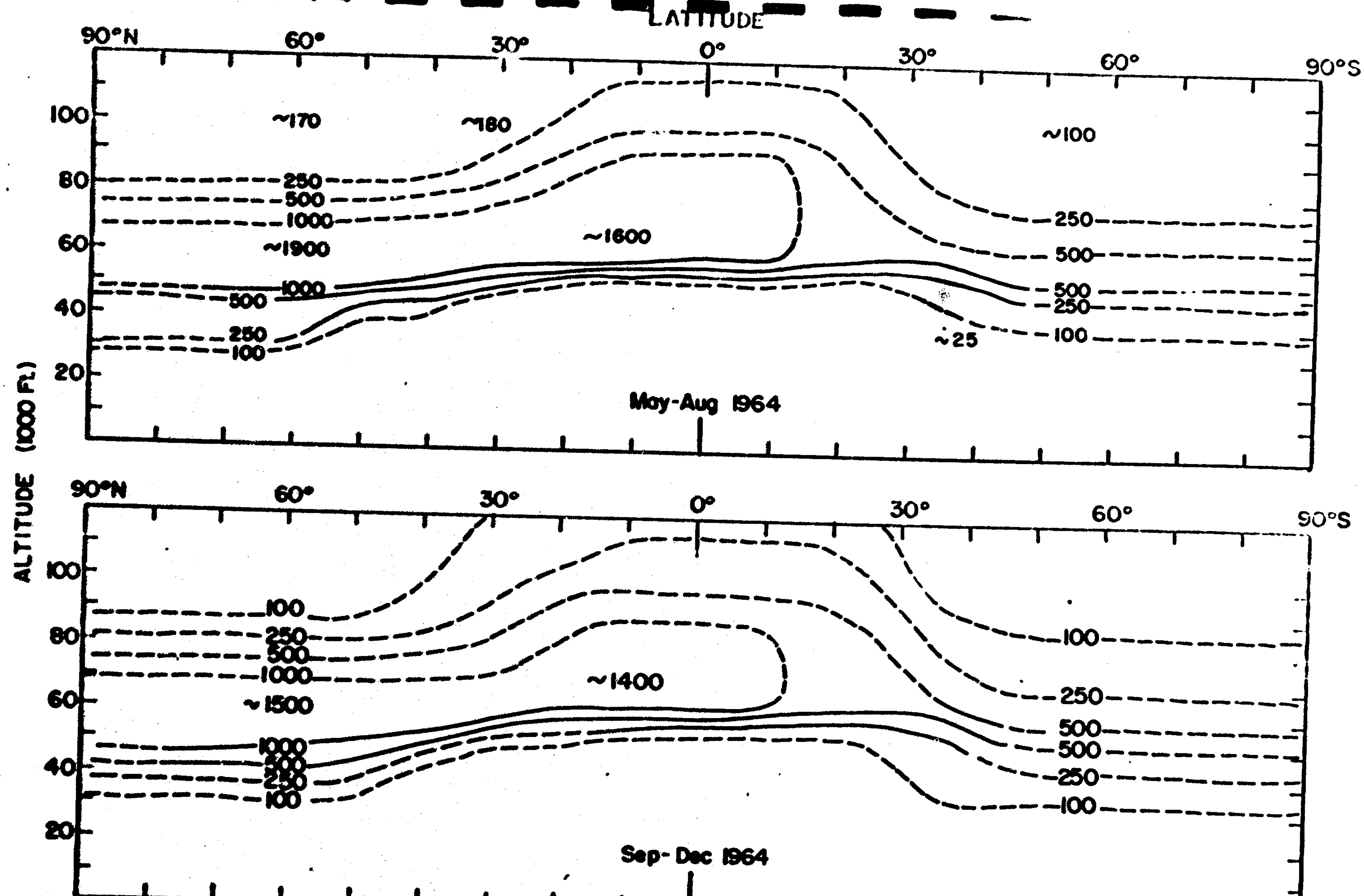


FIGURE 32. THE MEAN DISTRIBUTION OF MANGANESE-54 (dpm/1000 SCF corr. for decay to 31 Dec. 1962 DURING MAY-AUG 1964 AND SEP-DEC 1964

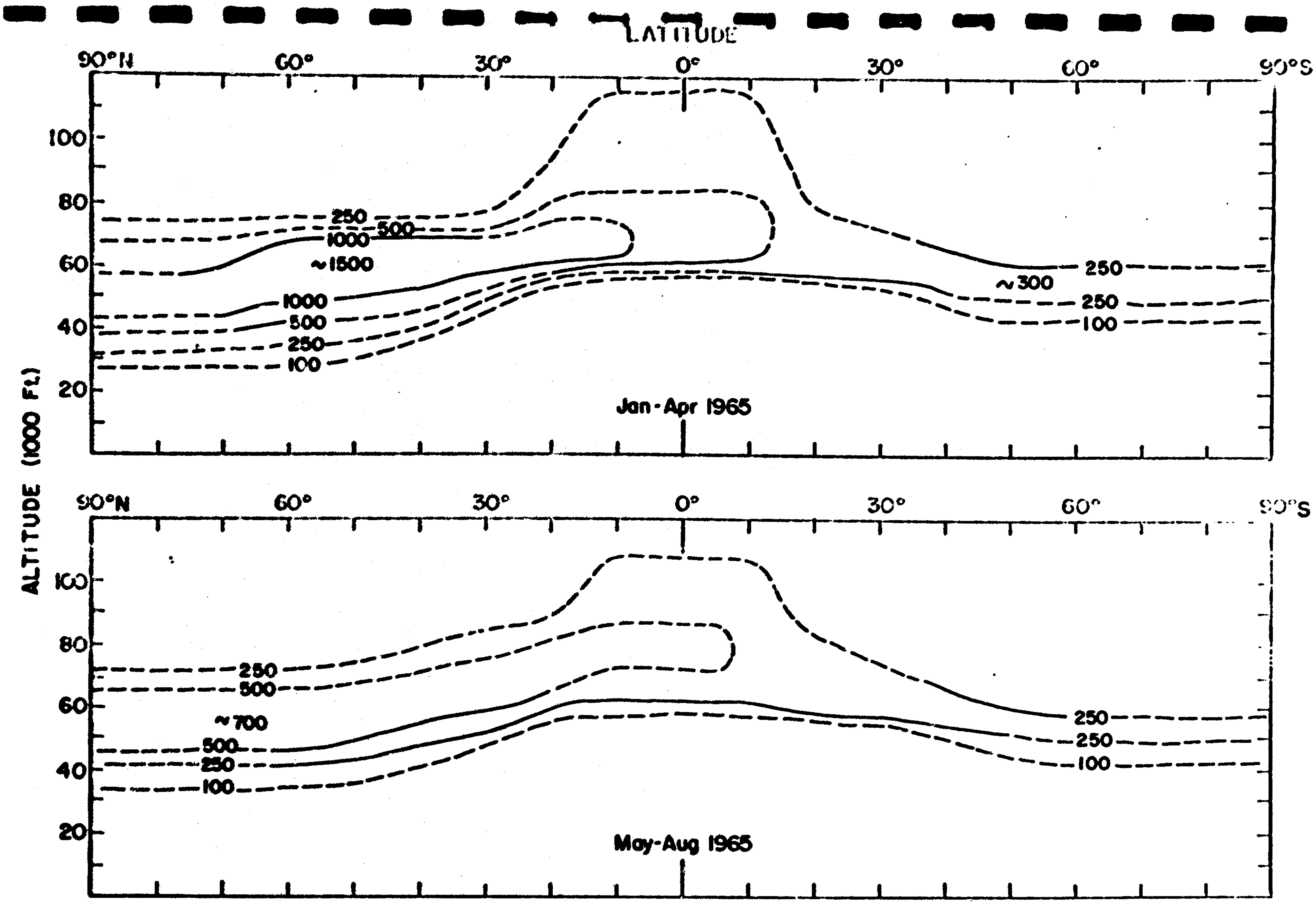


FIGURE 33. THE MEAN DISTRIBUTION OF MANGANESE-54 (dpm/1000 SCF corr. for decay to 31 Dec 1962) DURING JAN-APR 1965 AND MAY-AUG 1965

TABLE 3

THE STRATOSPHERIC BURDENS OF MANGANESE-54
(MEGACURIES CORRECTED FOR DECAY TO 31 DECEMBER 1962)

Time Interval	Northern Hemisphere	Southern Hemisphere	Total Burden
Jan - Apr 1963	22.8 ± 8.0	1.2 ± 0.8	24.0 ± 8.0
May - Aug 1963	13.6 ± 3.5	1.6 ± 1.0	15.2 ± 3.6
Sep - Dec 1963	9.4 ± 2.5	2.5 ± 1.2	11.9 ± 2.8
Jan - Apr 1964	7.7 ± 2.0	2.0 ± 1.0	9.7 ± 2.2
May - Aug 1964	4.2 ± 1.4	2.1 ± 1.0	6.3 ± 1.7
Sep - Dec 1964	3.7 ± 1.2	2.1 ± 1.0	5.8 ± 1.6
Jan - Apr 1965	3.4 ± 1.1	1.0 ± 0.5	4.4 ± 1.2
May - Aug 1965	2.0 ± 0.9	1.0 ± 0.5	3.0 ± 1.0

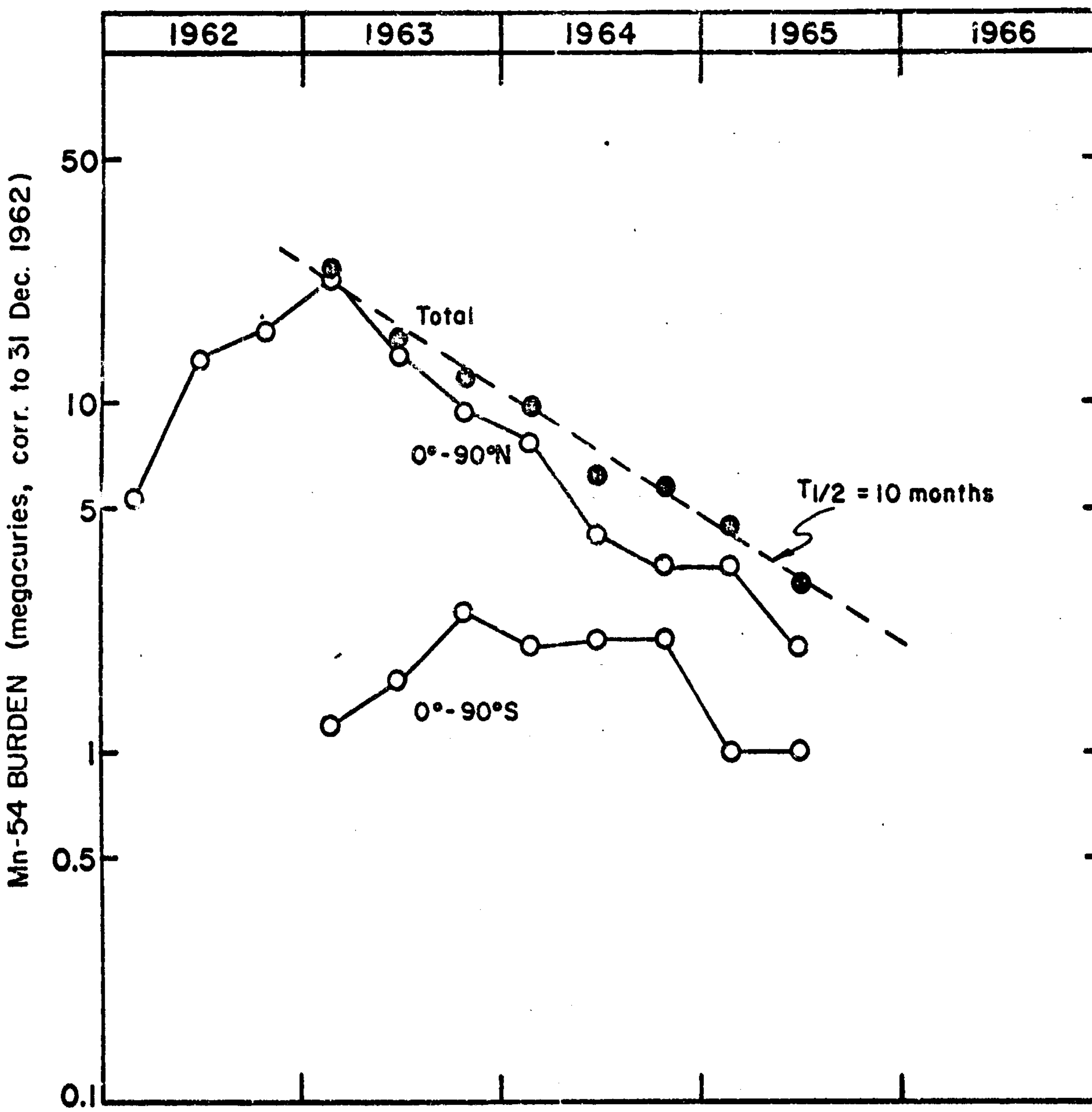


FIGURE 34. STRATOSPHERIC BURDENS OF MANGANESE-54

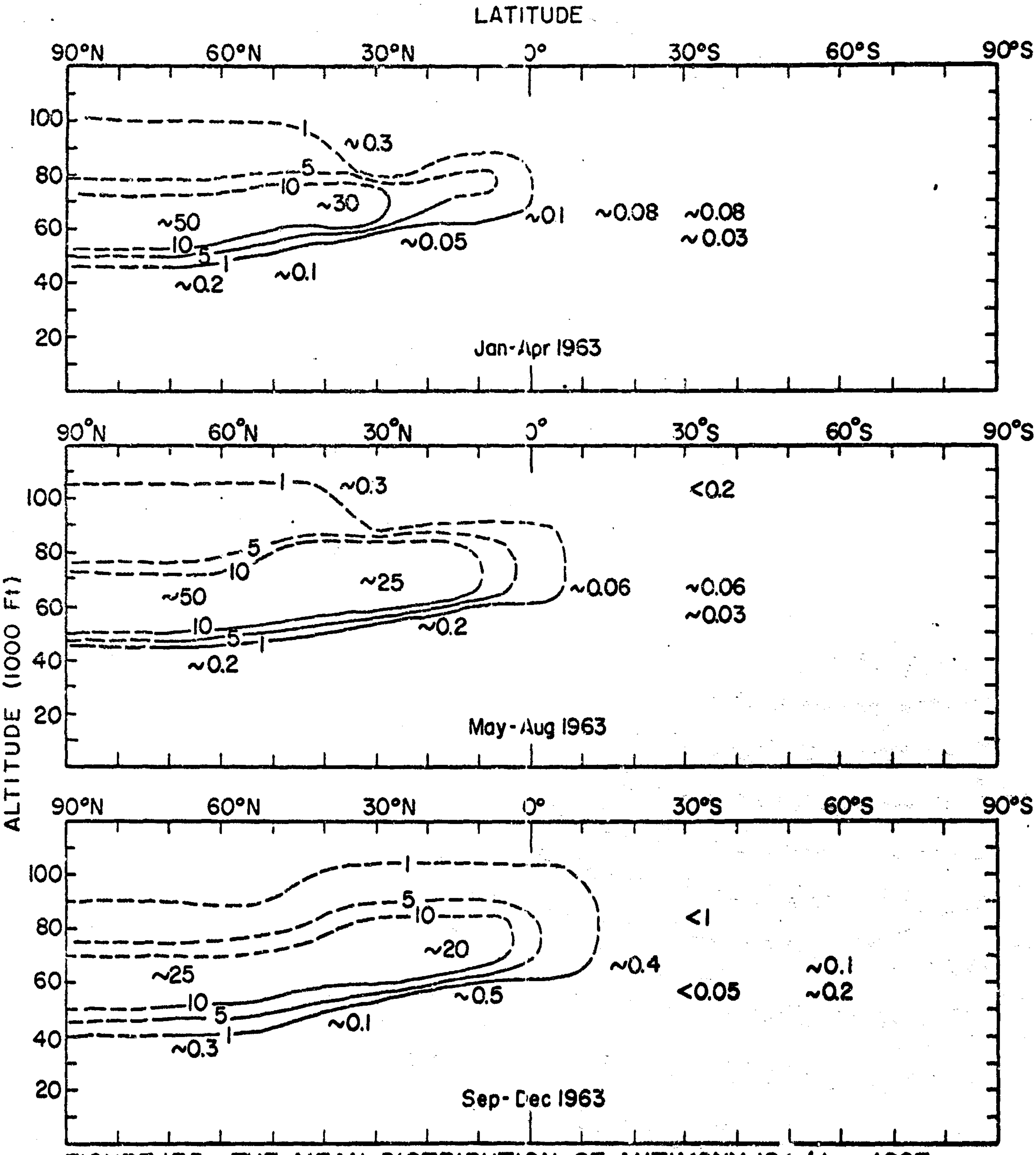


FIGURE 35. THE MEAN DISTRIBUTION OF ANTIMONY-124 (dpm / SCF corr. to 31 Dec 1962) DURING JAN-APR 1963, MAY-AUG 1963 AND SEP-DEC 1963

The Stratospheric Distribution of Yttrium-88

A 1.85 Mev peak attributable to yttrium-88 was found by Salter in gamma spectra of balloon samples collected at 31°N in late 1962 and in 1963. Subsequently this peak was also found in gamma spectra of some Stardust filter samples collected in the Northern Hemisphere during 1963. Judging from its time of appearance in stratospheric filter samples, the bulk of the yttrium-88 measured must have been produced by one or more events in the late 1962 nuclear weapons test series.

The stratospheric distributions of yttrium-88 during January-April 1963 and May-August 1963 are shown in Figure 36, and during September-December 1963 and January-April 1964 in Figure 37. Although the 1.85 Mev peak of yttrium-88 was observable in the gamma spectra of many USAEC balloon filter samples, the radiochemical analyses of these samples for this nuclide were not always successful. As a result the yttrium-88 concentrations in the upper stratosphere during 1963 and early 1964 are not well known. It was also difficult to obtain good data for samples collected in the lower stratosphere when their yttrium-88 activities, corrected for decay to 15 November 1962, were less than about 250 dpm/1000 SCF. It was only during September-December 1963 that activities in the lower stratosphere of the Southern Hemisphere clearly exceeded this value. By mid-1964 the continued radioactive decay of the yttrium-88 had reduced its concentrations to values below the limit of detection in much of the stratosphere even of the Northern Hemisphere, and measurements were discontinued.

Concentrations of yttrium-88, as of other particulate radioactive debris, in the lower stratosphere of the Northern Hemisphere decreased during 1963-1964 as a result of fallout and of dilution due to lateral and vertical mixing within the stratosphere. Significant quantities of yttrium-88, as well as of strontium-90 and manganese-54, were transferred into the Southern Hemisphere during the winter-spring of 1963. Thus the behavior of yttrium-88 in the stratosphere during 1963 to early 1964 was similar to that of other debris from the late 1962 nuclear weapons tests.

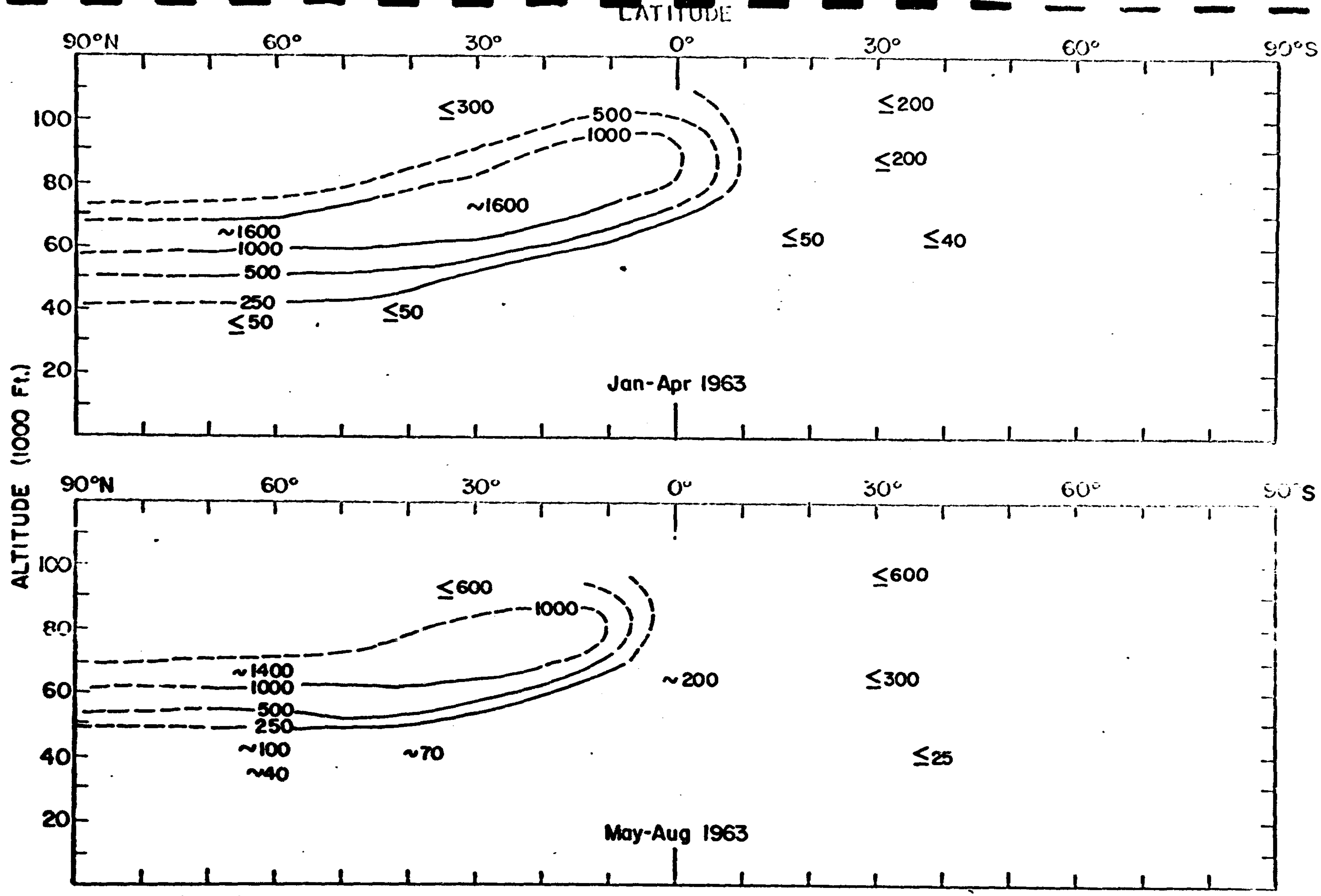


FIGURE 36. THE MEAN DISTRIBUTION OF YTTRIUM-88 (dpm/1000 SCF corr for decay to 15 NOV 1962) DURING JAN-APR 1963 AND MAY-AUG 1963

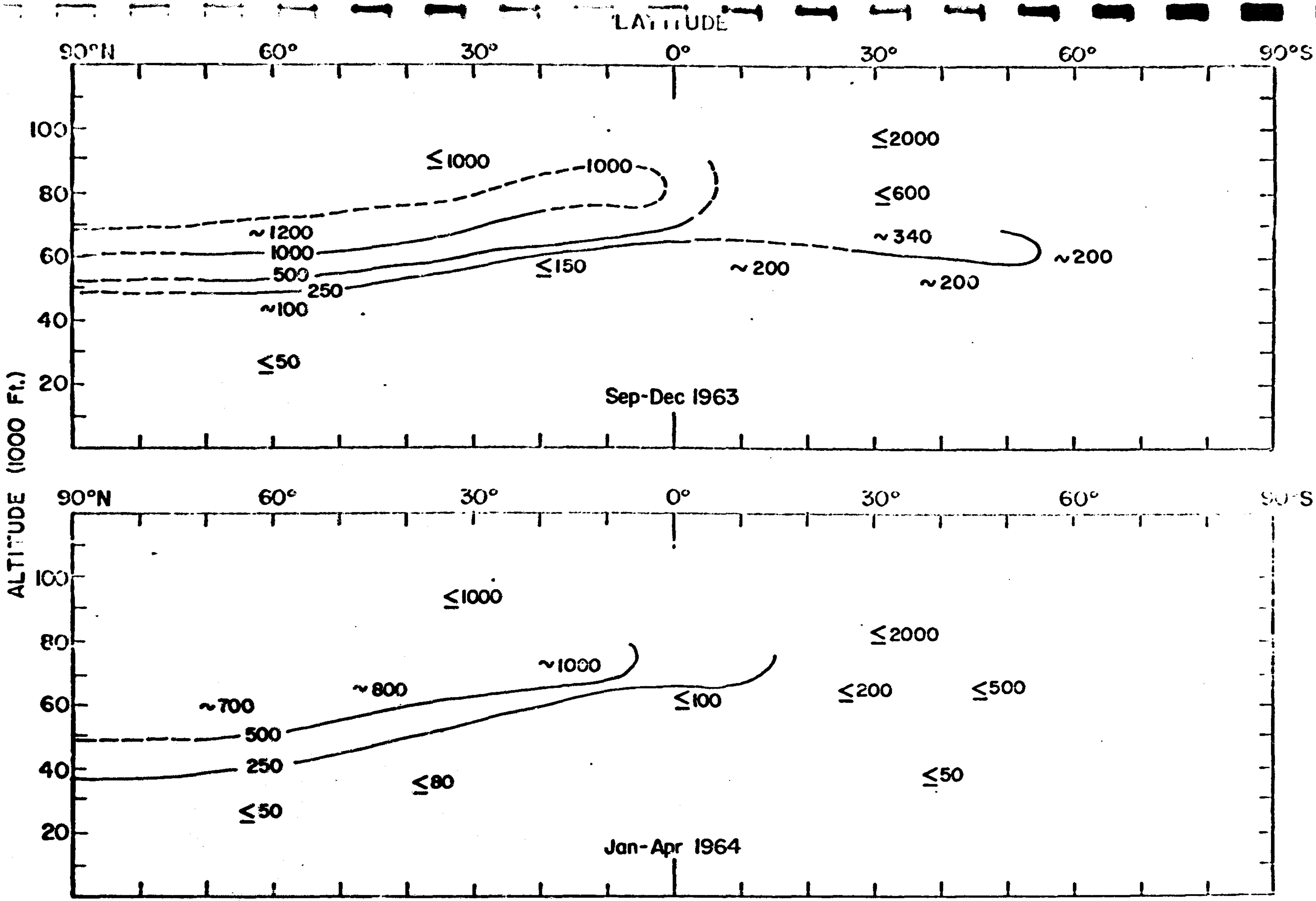


FIGURE 37. THE MEAN DISTRIBUTION OF YTTRIUM-88 (dpm/1000 SCF corr. for decay to 15 NOV 1962) DURING SEP-DEC 1963 AND JAN-APR 1964

The Stratospheric Distribution of Artificial Sodium-22

Beginning in late 1963 a series of Stardust samples was analyzed for several products of cosmic ray activity, including sodium-22. It was found that the sodium-22 concentrations in the lower stratosphere of the Northern Hemisphere far exceeded the expected equillibrium concentrations of natural sodium-22. The expected distribution of natural sodium-22 in the Stardust sampling corridor, assuming the atmosphere to be stagnant and the rate of decay of sodium-22 at each point to be equal to its rate of production, is shown in the upper half of Figure 38. The discontinuity in the isolines between 10°S and 15°S results from the difference in longitude and, therefore, of geomagnetic latitude, between sampling missions flown southward from the Canal Zone and northward across Australia.

The observed distribution of sodium-22 in the Stardust sampling corridor during January-March 1964 is shown in the lower half of Figure 38. In the southern polar stratosphere concentrations are similar to those expected, but in the northern polar stratosphere and in the tropical stratosphere they are considerably higher than expected. These high stratospheric concentrations, as well as high concentrations of sodium-22 found in rainfall during 1963 and 1964[7], are evidence of the artificial production of this nuclide by the 1961 and 1962 nuclear weapons test series[8].

Sodium-22 was measured in a series of samples collected during 1961-1963 in an attempt to establish the general time of origin of the sodium-22. The sodium-22 concentration in three samples collected during July, 1961 and in one collected on 6 September 1961, before the first interception by Stardust of debris from the 1961 U.S.S.R. weapons test series, ranged from 0.3 to 1.2 dpm/1000 SCF. When debris from the U.S.S.R. tests was intercepted, concentrations ranging from below 1.0 up to about 30 dpm/1000 SCF were found. There is a similarity between the pattern of occurrence of high sodium-22 concentrations and high iron-55,

manganese-54 and antimony-124 concentrations, which suggests that all of these activation products had the same source.

Further increases in the stratospheric concentrations of sodium-22 occurred during late 1962, no doubt as a result of the 1962 series of tests of nuclear weapons. Trends in sodium-22 concentrations at 50 and 65 thousand feet at 60°-70°N and at 65 thousand feet at 40°-45°N and 0°-10°N are shown in Figures 39 and 40. Data reported by Bhandari et al[8] are combined with Stardust data in these figures. The Stardust data for all three latitudes show a peak concentration in early 1963, in samples containing debris from the late 1962 weapons test series. The data of Bhandari et al show a peak in late 1963 at all three latitudes. Available Stardust measurements provide some slight confirmation of these late 1963 peaks, but no similar peaks are found in data for other activation products which were being measured during late 1963. As is pointed out by Bhandari et al, the late 1963 peaks in sodium-22 concentration, appearing as they do about a year or so after the injection of the nuclide, may be a very significant observation of a feature which had previously not been reported. It is possible, however, that they may be analagous to peaks in rhodium-102 or cadmium-109 concentrations which were observed relatively long after the injections of these nuclides because these injections took place at high altitudes. Bhandari et al believe that peaks in sodium-22 concentrations in late 1963 may represent the arrival in the lower stratosphere of sodium-22 from a high altitude injection.

The sodium-22 data may be applicable to studies of rates of interhemispheric transfer of debris, although this potential usefulness is limited by some inconsistencies in the data which may or may not have resulted from analytical errors. In Figure 41 are plotted the sodium-22 activities encountered at 60 and at 65 thousand feet during each of six months: February, May and September 1963 and February, July and September 1964. Again data published by Bhandari et al[8] as well

as Stardust data have been plotted. In February 1963 the concentration gradient across the tropical stratosphere was very steep, with concentrations in the northern polar stratosphere being more than ten times as high as those in the southern polar stratosphere. The gradient grew less steep during the course of 1963 and 1964, and by September, 1964 quite similar concentrations were found in the two polar stratospheres.

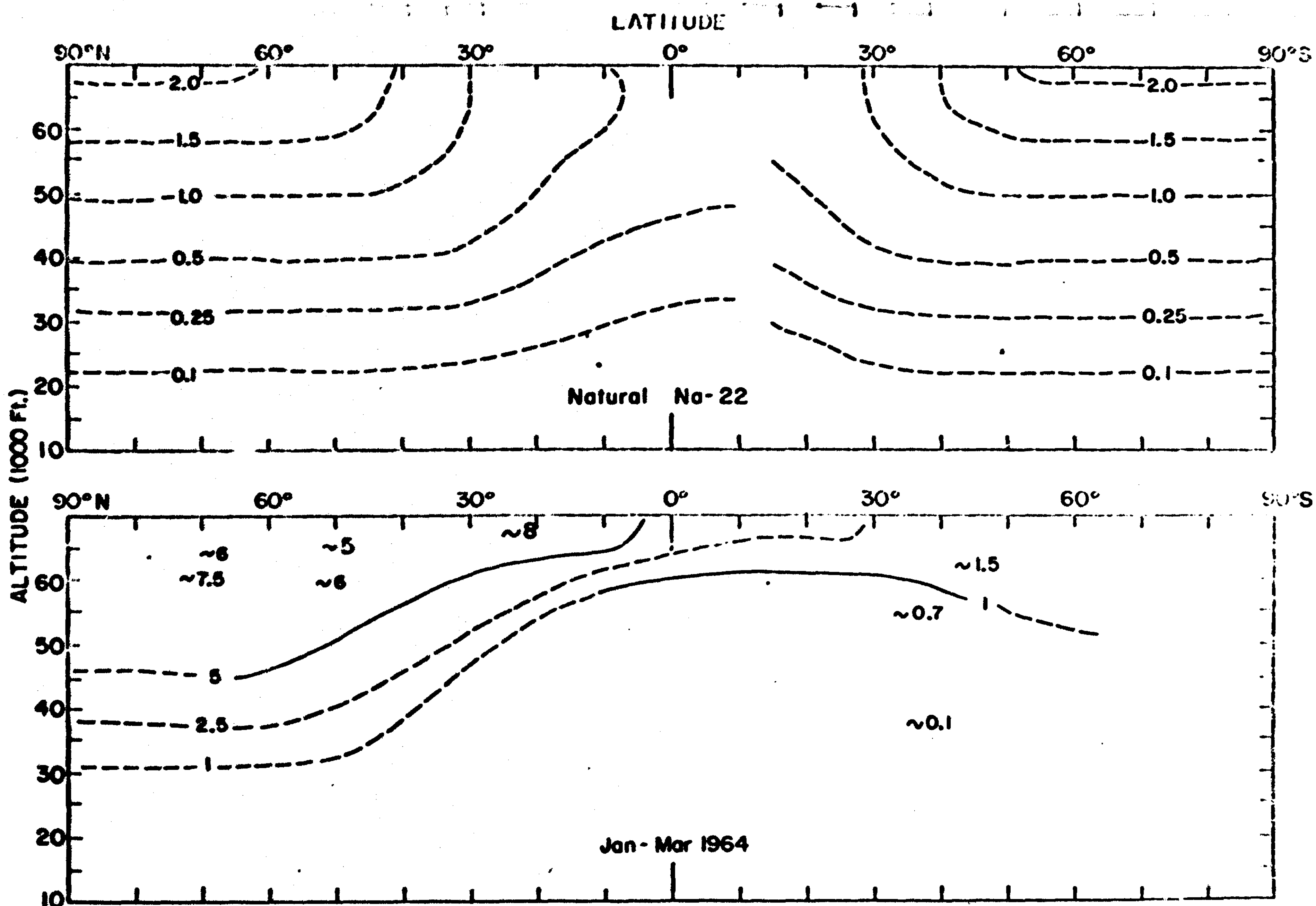


FIGURE 38. EQUILIBRIUM CONCENTRATIONS OF NATURAL SODIUM-22 IN THE STARDUST SAMPLING CORRIDOR AND THE MEAN DISTRIBUTION OF SODIUM-22 DURING JANUARY - MARCH 1964

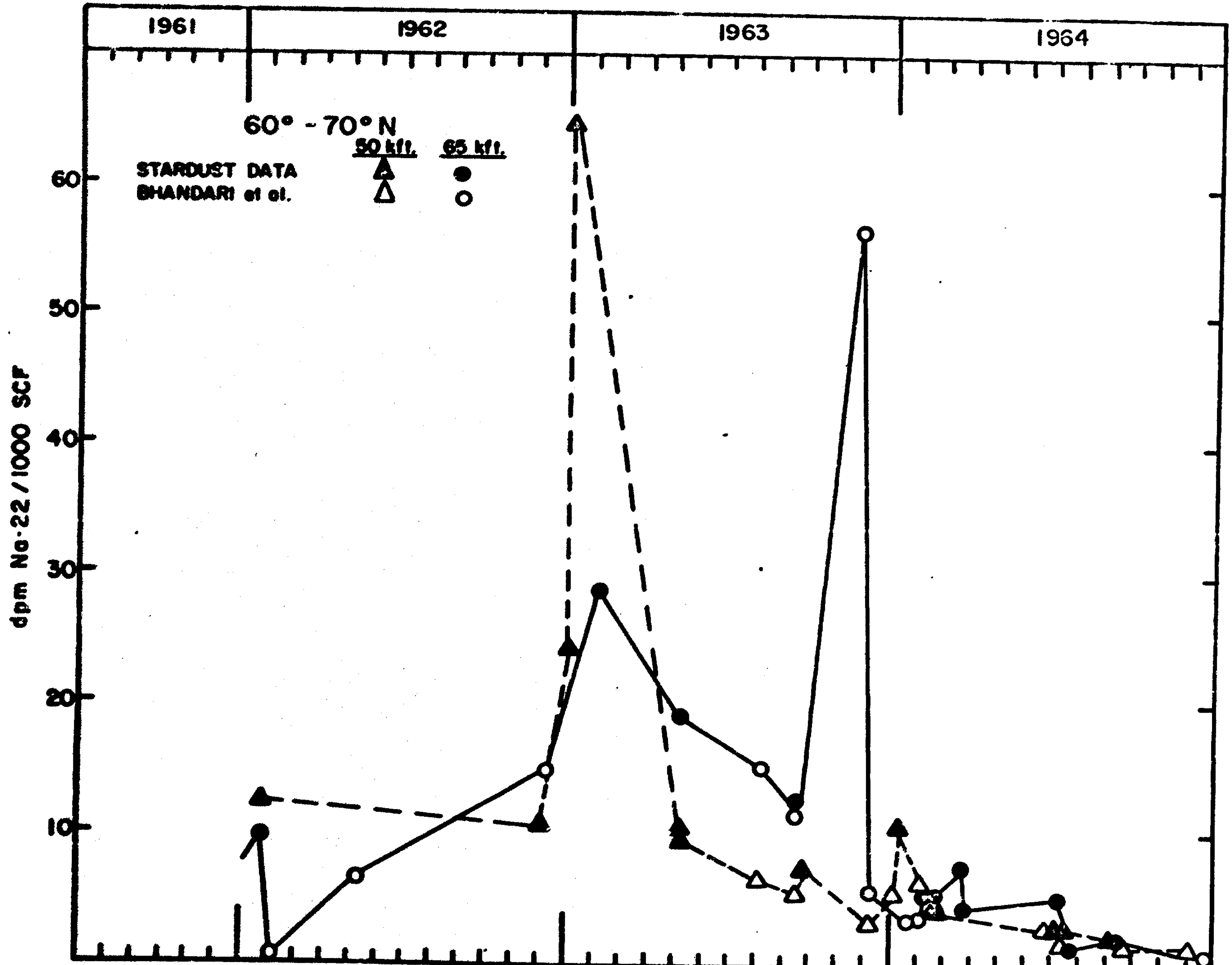


FIGURE 39. SODIUM-22 CONCENTRATIONS AT 60° - 70° N

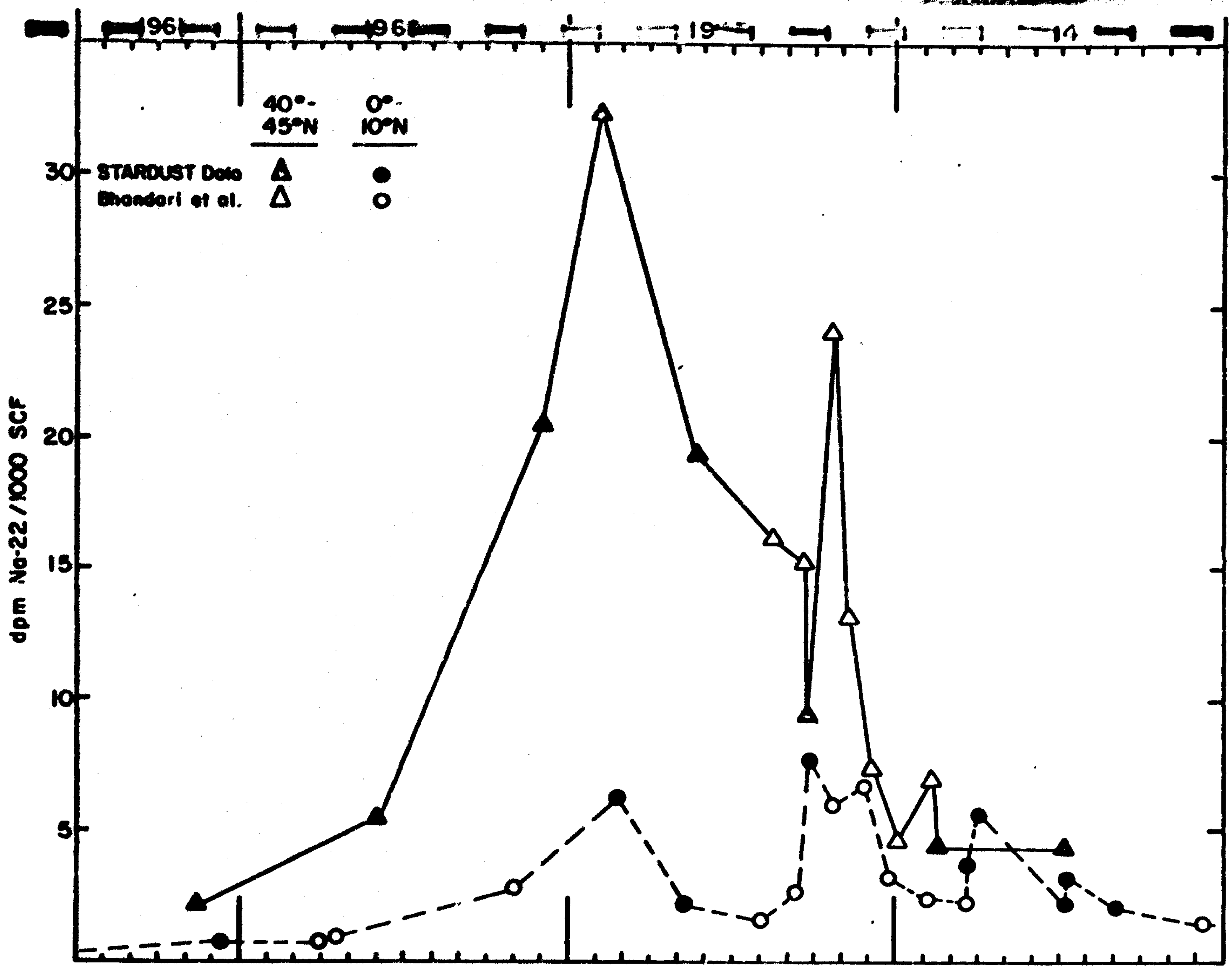


FIGURE 40. SODIUM-22 CONCENTRATIONS AT ABOUT 65,000 FEET AT 40°-45°N AND AT 0° - 10°N

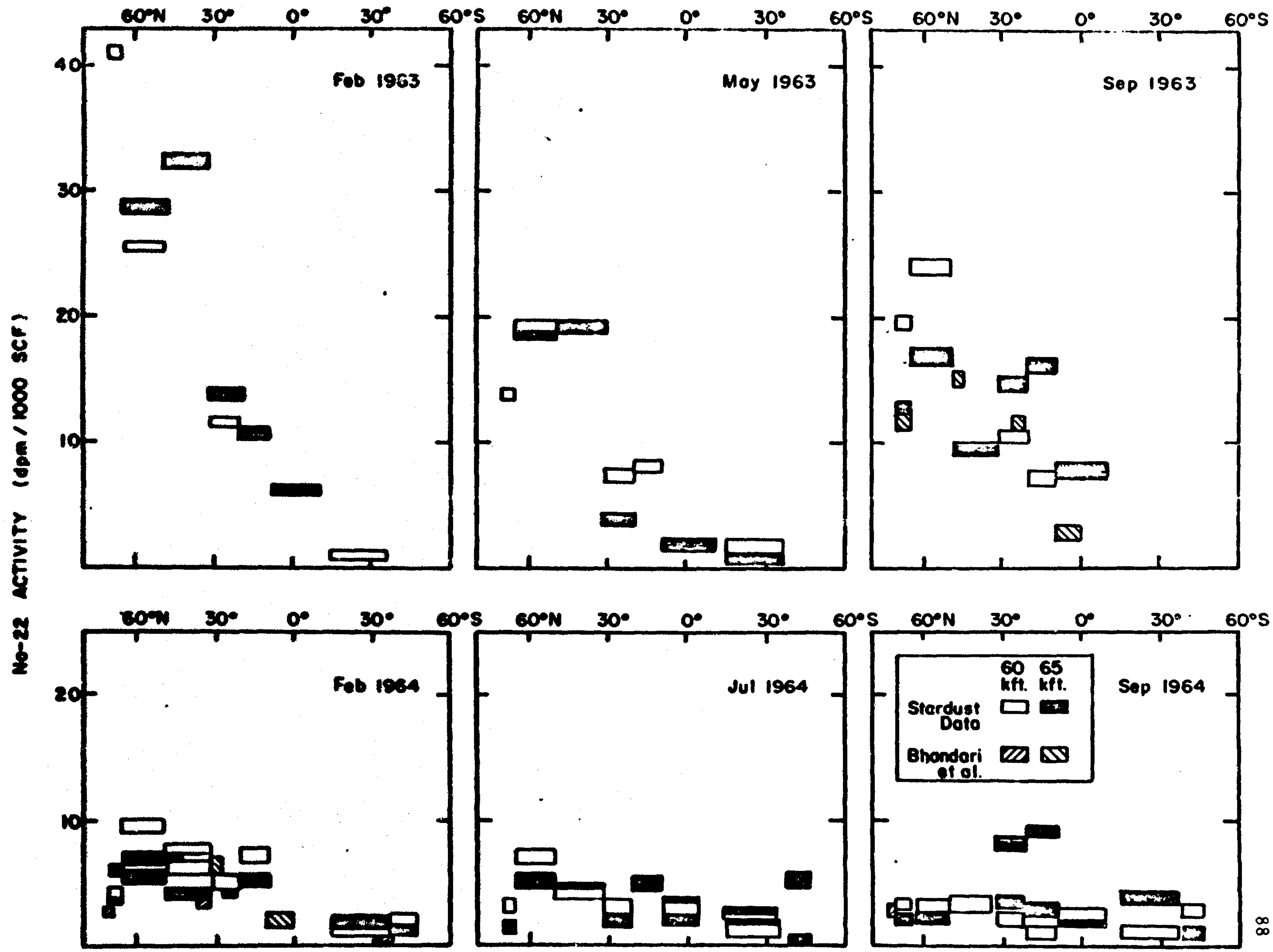


FIGURE 41 · LATITUDINAL DISTRIBUTION OF SODIUM-22 ACTIVITY AT 60,000 AND 65,000 FT.

The Stratospheric Distribution of Cadmium-109

It has been estimated that between 100 and 400 kilocuries of cadmium-109 were produced by the Starfish Prime event of the Dominic series of nuclear weapons tests[9,10]. This event, which took place at a height of about 400 km in the vicinity of Johnston Island (about 17°N latitude) on 9 July 1962, had a reported yield of 1.4 megatons. Much of the cadmium-109 was, no doubt, injected into the upper atmosphere by this event, but some may have been ejected from the atmosphere. Fission fragments in the debris would be self-ionizing and would be trapped in the magnetosphere, but this may not have been true of the non-fission fragment components containing the cadmium[10]. Thus, less than 100 kilocuries of cadmium-109 may have remained in the atmosphere following the explosion of the Starfish Prime device.

Cadmium-109 from the Starfish Prime event was first detected in the stratosphere by the USAEC high altitude balloon sampling program. A sample collected at 105 thousand feet in the vicinity of Mildura, Australia (34°S latitude) on 13 December 1962 contained a high concentration of this nuclide. Concentrations of cadmium-109 found at about 105 and 78 thousand feet near Mildura, and at about 105 and 80 thousand feet at San Angelo, Texas (31°N) during the AEC high altitude balloon program[2,3,4] are plotted in Figure 42.

At 105 thousand feet at 34°S, subsequent to the first interception of cadmium-109 in December 1962, still higher concentrations were intercepted during March and April, 1963. In May and July, 1963 samples collected at this location contained virtually no cadmium-109, however, indicating that this tracer nuclide was still far from being distributed uniformly within the upper stratosphere of the Southern Hemisphere. All samples collected at this location between August, 1963 and late 1965 contained significant cadmium-109 activities, but there was a considerable decrease in the average concentrations between late 1963 and late 1964, and again between late 1964 and late 1965. At 78 thousand feet at 34°S

cadmium-109 concentrations increased during late 1963 and, during the first two thirds of 1964, were consistently higher than the concentrations at 105 thousand feet at that latitude. During late 1964 and during 1965, however, comparable concentrations were found at both levels.

Cadmium-109 concentrations at 105 and 80 thousand feet at 31°N underwent a significant increase during 1964, with concentrations at 105 thousand feet at this latitude reaching higher values than were present at a comparable altitude at 34°S. By 1965, however, concentrations at both of these altitudes were similar to each other and to those found at comparable altitudes at 34°S.

Cadmium-109 data for the lower stratosphere, obtained by analysis of Stardust filter samples, also indicate that during the past few years a gradual equalization of concentrations between the Northern and Southern Hemispheres has taken place. In Figure 43 are plotted results for samples collected at 65 thousand feet at 25°S, at 55 and 65 thousand feet at 45°S, and at 55 and 65 thousand feet at 65°N, during 1962 to 1965.

Cadmium-109 from the Starfish Prime event first reached the lower stratosphere towards the end of 1963 in the Southern Hemisphere, and at about the beginning of 1964 in the Northern. Low concentrations of cadmium-109, presumably produced by some events in the 1962 U.S.S.R. weapons tests series, were found in the Northern Hemisphere during late 1962 and early 1963. During 1964 the concentrations found in the Southern Hemisphere far exceeded those found in the Northern, but by mid-1965 activity levels in the Southern Hemisphere had decreased significantly, and less difference remained between the activities in the two hemispheres.

Observed changes in the vertical profiles of cadmium-109 concentrations suggest that the vertical distribution of this tracer, as well as its distribution between hemispheres, was becoming more uniform during 1963 to 1965. Vertical profiles for 1963, 1964, and 1965 obtained by combining data from the AEC balloon

program and from Project Stardust, are shown in Figure 44. At 34°S the highest concentrations at first were found at 110 thousand feet, and the concentration profile between 110 and 70 thousand feet was very steep in April, 1963, less steep in September, 1963, and, apparently, reversed in direction by March, 1964, with the highest concentration by then occurring at 70 thousand feet. By October-November, 1965 concentrations were much lower, and the maximum was found at 53 thousand feet. At 31°N the concentrations were low and the profile quite gentle in March, 1963 and October, 1963. The concentrations were considerably higher in March, 1964, with the maximum at the highest altitude sampled, but the profile was still gentle. By July, 1965 concentrations had decreased at the higher altitudes and the maximum was found at 65 thousand feet.

Vertical profiles of cadmium-109 activity in the lower stratosphere during 1963 to 1965, as deduced from Project Stardust data, are shown in Figures 45 and 46. At 40°S (Figure 45), both the concentrations and the steepness of the gradient between 65 and 55 thousand feet increased from May, 1963 to November, 1963 to April, 1964. By December, 1964, however, concentrations had decreased at 65 thousand feet. By September, 1965 they had decreased still further at that altitude, and had decreased at the lower altitudes as well. During both December, 1964 and September, 1965 a maximum in the vertical profile was found in the lower stratosphere, below 65 thousand feet.

At 65°N (Figure 46) some cadmium-109 was present in the lower stratosphere during January 1963, before the cadmium-109 from the Starfish Prime event had reached even the regions of the upper stratosphere sampled by the AEC balloon program. Perhaps a small amount of this nuclide was produced by one or more low altitude nuclear weapons tests during late 1962. The cadmium-109 concentrations in the lower northern polar stratosphere decreased rapidly during the first half of 1963, soon reaching values near the lower limit of detection of this nuclide. In any case, in June, 1963 there was no detectable cadmium-109 attributable to the Starfish Prime

event present in the lower stratosphere of the Northern Hemisphere. By February, 1964, however, cadmium-109 from this source had reached the 65 thousand feet level at this latitude. By January, 1965 there had been a considerable increase in concentrations above the 40 thousand feet level, and a steep concentration gradient had been established between 63 thousand feet and 40 thousand feet. In September, 1965 a steep gradient still existed between 60 thousand and 43 thousand feet, but very similar cadmium-109 activities were found at 60 and at 65 thousand feet.

The vertical profiles in the Northern Hemisphere (Figures 44 and 46) indicate that during 1963 to 1965 significant quantities of this tracer were appearing at lower and lower altitudes. Presumably then, the vertical distribution of cadmium-109 was becoming progressively more uniform during these years, especially if the observed downward movement resulted from eddy diffusion. The vertical profiles in the Southern Hemisphere (Figures 44 and 45) indicate that there, this movement progressed much more rapidly than it did in the Northern Hemisphere. They also suggest that by the end of December, 1964 there existed a fairly uniform vertical distribution of cadmium-109 between the upper stratosphere and levels as low as 55 thousand feet in the lower stratosphere, and, further, that a layer of maximum concentration may have begun to form at about 60 thousand feet. If this latter observation is confirmed as additional evidence is accumulated, the most likely explanation of this phenomenon would be in terms of the combined action of vertical eddy diffusion and particle settling.

By combining cadmium-109 data from Project Stardust and the AEC balloon program it is possible to estimate the approximate distribution of this nuclide in the stratosphere during 1964 and 1965. The available data seem to be consistent with the concept that by late 1964 a fairly uniform vertical distribution of cadmium-109 existed between the upper stratosphere and the higher layers of the lower stratosphere. Estimates have been made of the stratospheric distribution of cadmium-109 during several intervals in 1964 and 1965. The distributions for January

to April and May to August 1965 are shown in Figure 47. On the basis of these estimated distributions, calculations have been made of the stratospheric burden of cadmium-109 during 1964 and during the first two-thirds of 1965. The results of these calculations are summarized in Figure 48. They suggest that during 1964 and the first third of 1965 there were approximately 70 kilocuries of cadmium-109 in the stratosphere and upper atmosphere, but that by mid-1965 this total had decreased considerably, presumably mainly as a result of fallout to the troposphere in the Southern Hemisphere.

Of course, the uncertainties in the calculated burdens are large. As is seen from the upper half of Figure 48, the greater part of the cadmium-109 is in the Southern Hemisphere, where both the Stardust and balloon sampling are more restricted in latitudinal coverage than they are in the Northern Hemisphere. Indeed, Stardust sampling of the lowest layers of the southern polar stratosphere, between 53 thousand feet and the tropopause, is especially sparse, and by mid-1965 this region contained a very significant fraction of the total cadmium-109 burden. As is seen from the lower half of Figure 48, we estimate that the burdens of cadmium-109 in the unsampled region above 10 mb (about 100 thousand feet) and in the region between 10 and 50 mb, which is sampled by the balloon program, were decreasing steadily during late 1964 and during 1965. The accuracy of this estimate will be more easily judged when data for 1966 become available. The cadmium-109 concentrations in the entire stratosphere should continue to decrease during 1965-1966.

If our calculated stratospheric distributions of cadmium-109 are correct, we would expect that there was almost no fallout of Starfish Prime cadmium-109 before late 1964, about 30 months following its injection, but that since the beginning of 1965 this debris has fallen out at a rate equivalent to a stratospheric residence half time of 12 to 18 months. Apparently half of the cadmium-109 will have fallen out during the first 3.5 to 4.0 years following its injection, but three-

quarters may have fallen out by the time 4.5 to 5.5 years have elapsed since injection.

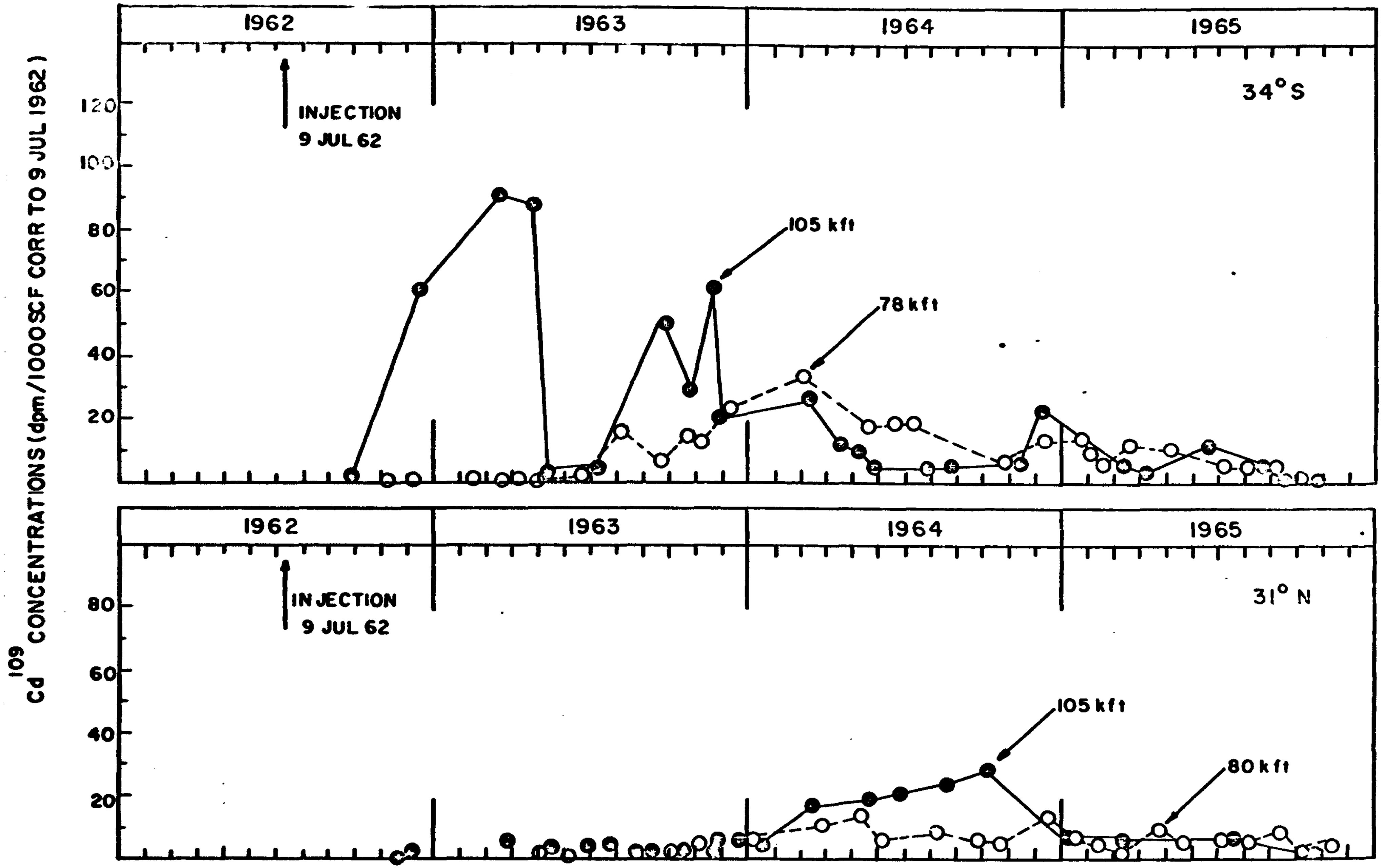


FIGURE 42. VARIATIONS WITH TIME IN CADMIUM-109 ACTIVITIES IN AEC BALLOON SAMPLES

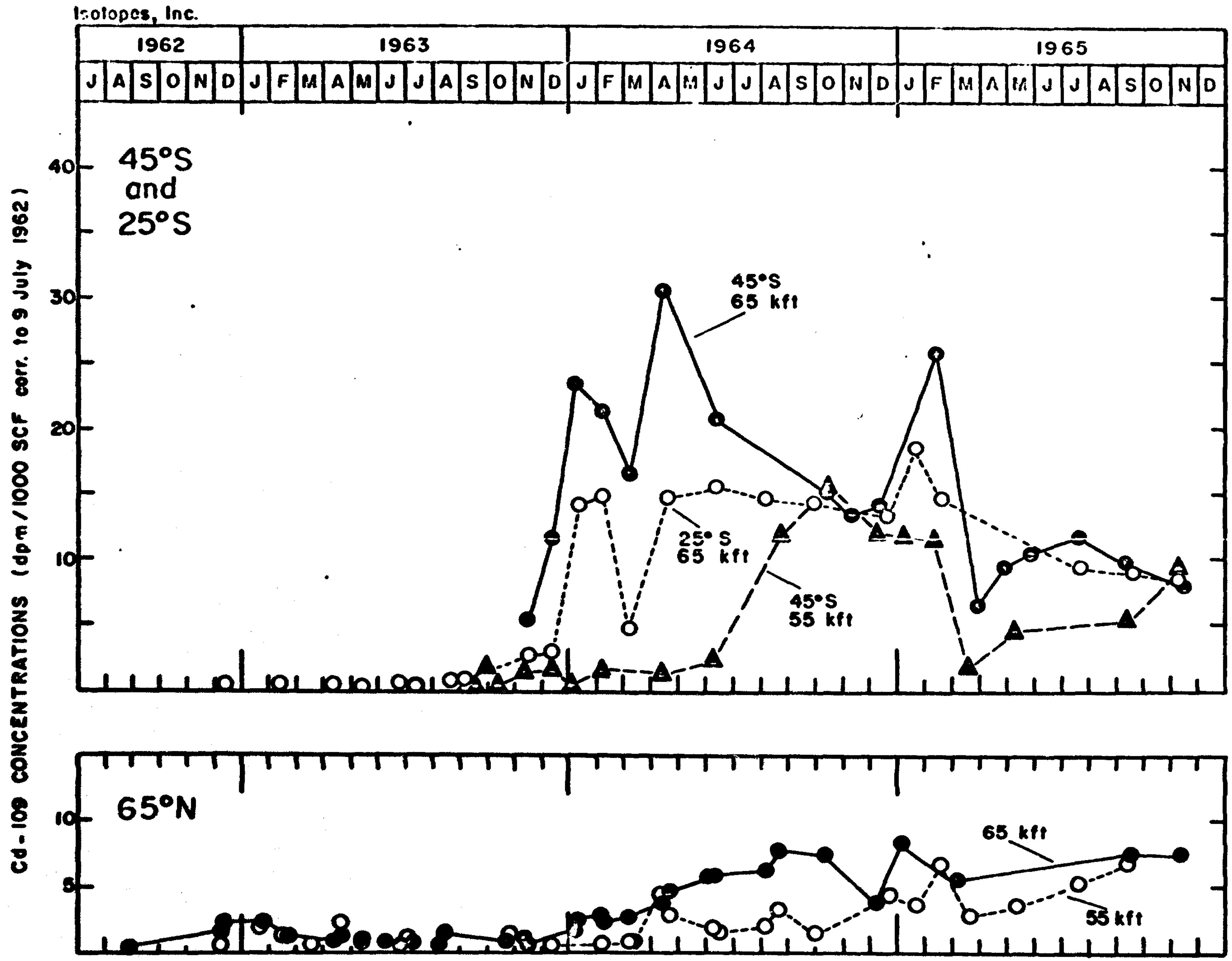


FIGURE 43. VARIATIONS WITH TIME IN CADMIUM-109 ACTIVITIES IN STARDUST SAMPLES

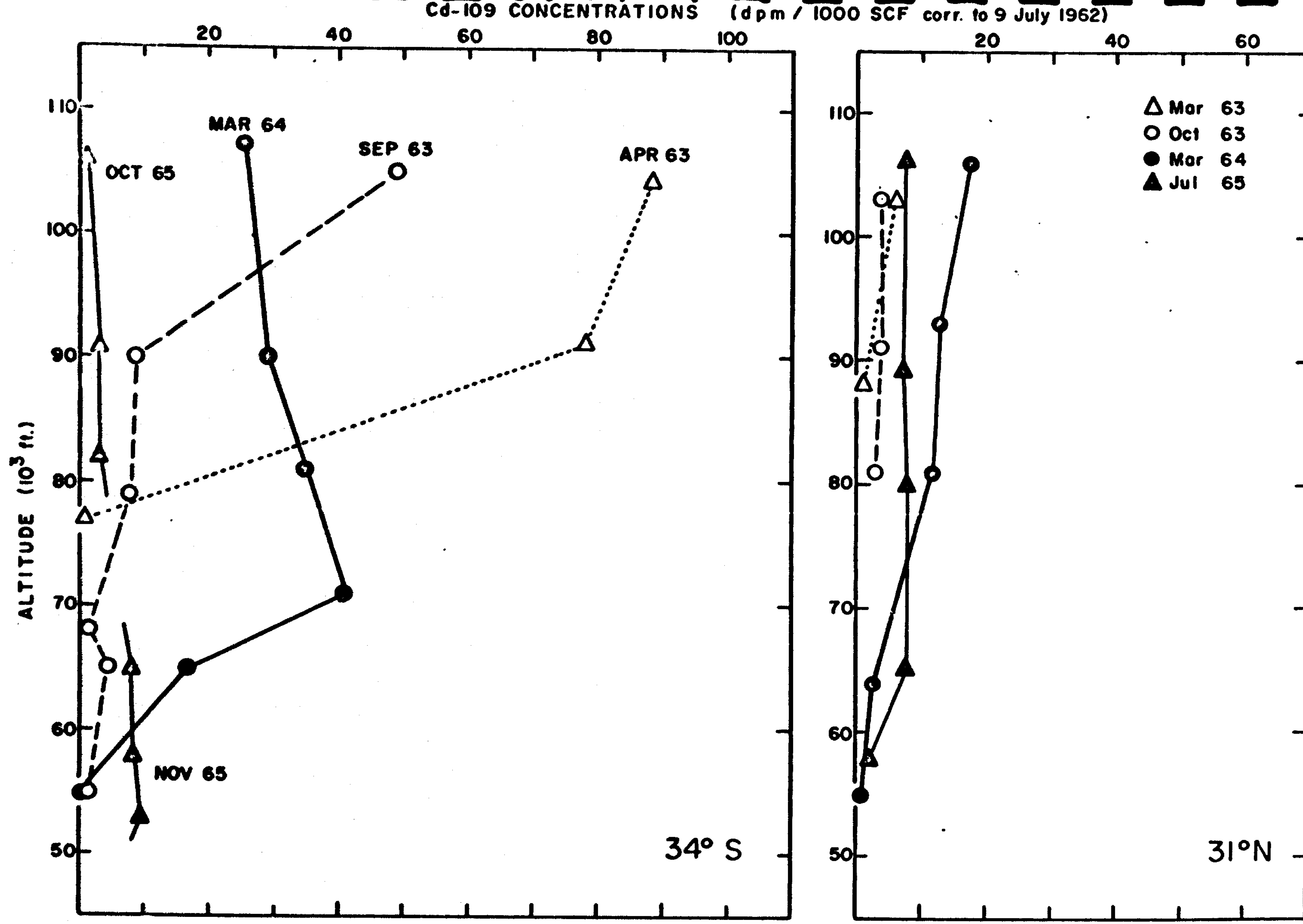


FIGURE 44. VERTICAL PROFILES OF CADMIUM-109 ACTIVITIES AT 34°S AND 31°N

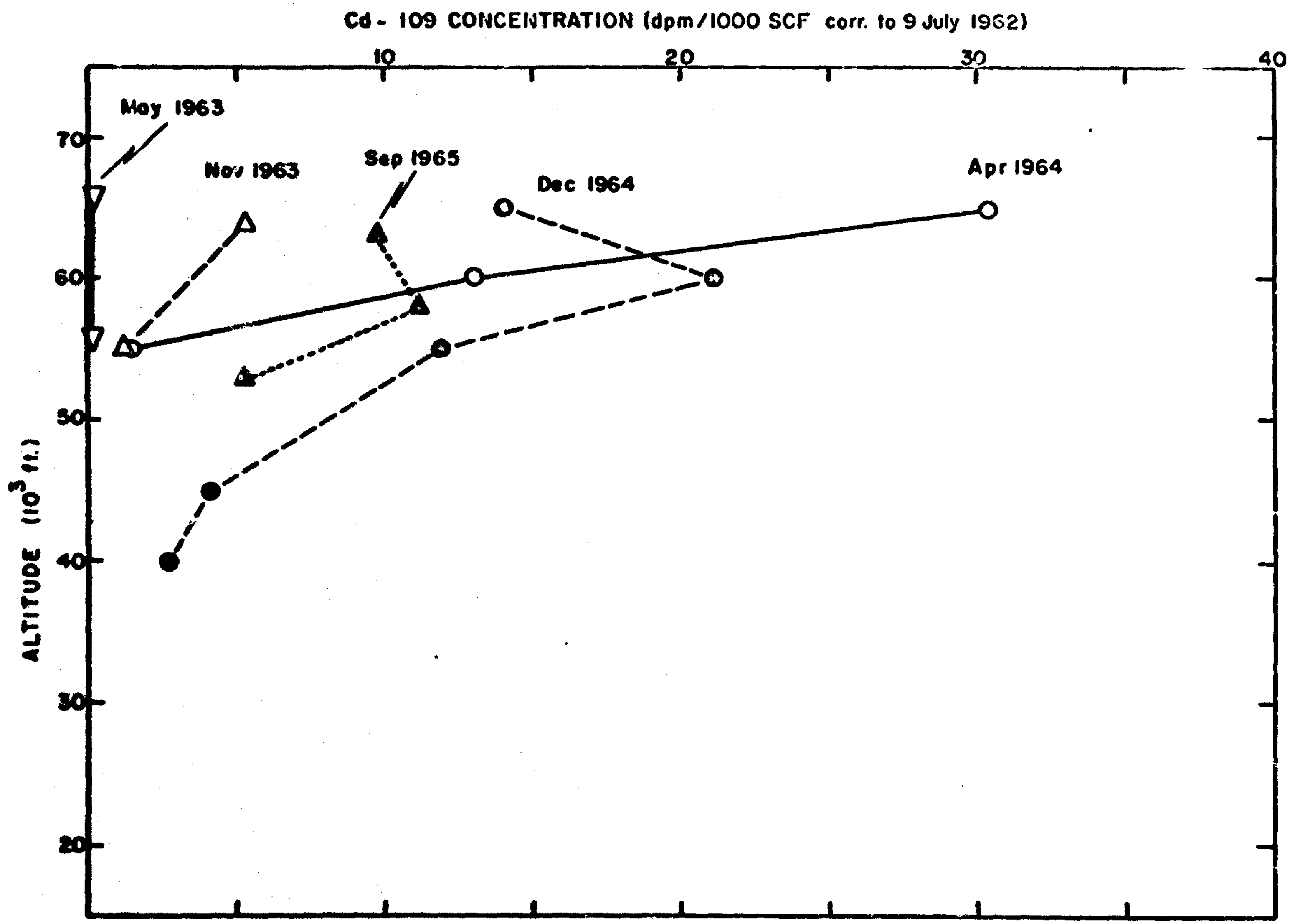


FIGURE 45 VERTICAL PROFILES OF CADMIUM-109 AT 40°S IN THE LOWER STRATOSPHERE

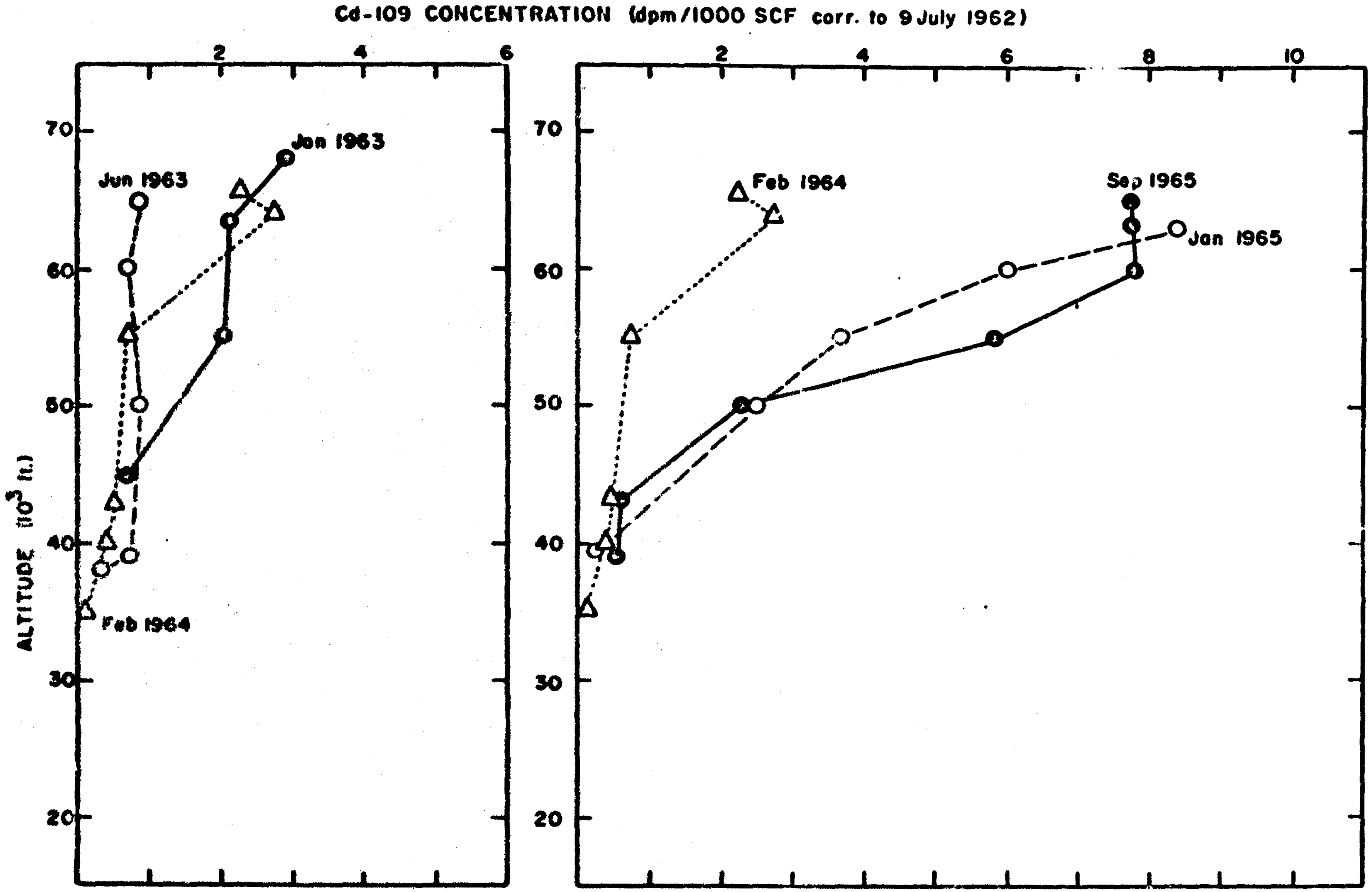


FIGURE 46. VERTICAL PROFILES OF CADMIUM-109 AT 65°N, IN THE LOWER STRATOSPHERE

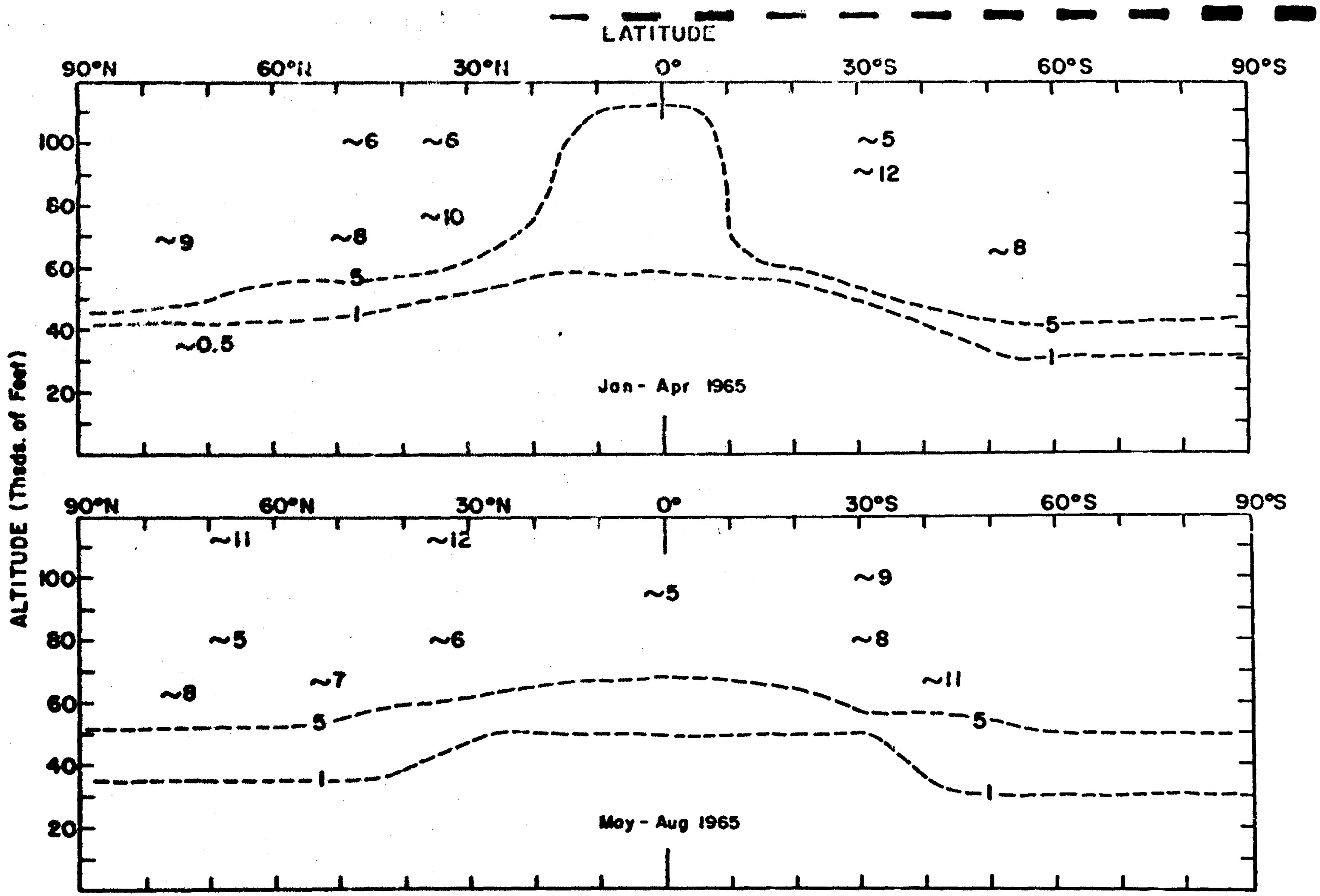


FIGURE 47. THE MEAN DISTRIBUTION OF CADMIUM-109 (dpm/1000 SCF corrected for decay to 9 July 1962) DURING JANUARY-APRIL AND MAY-AUGUST 1965

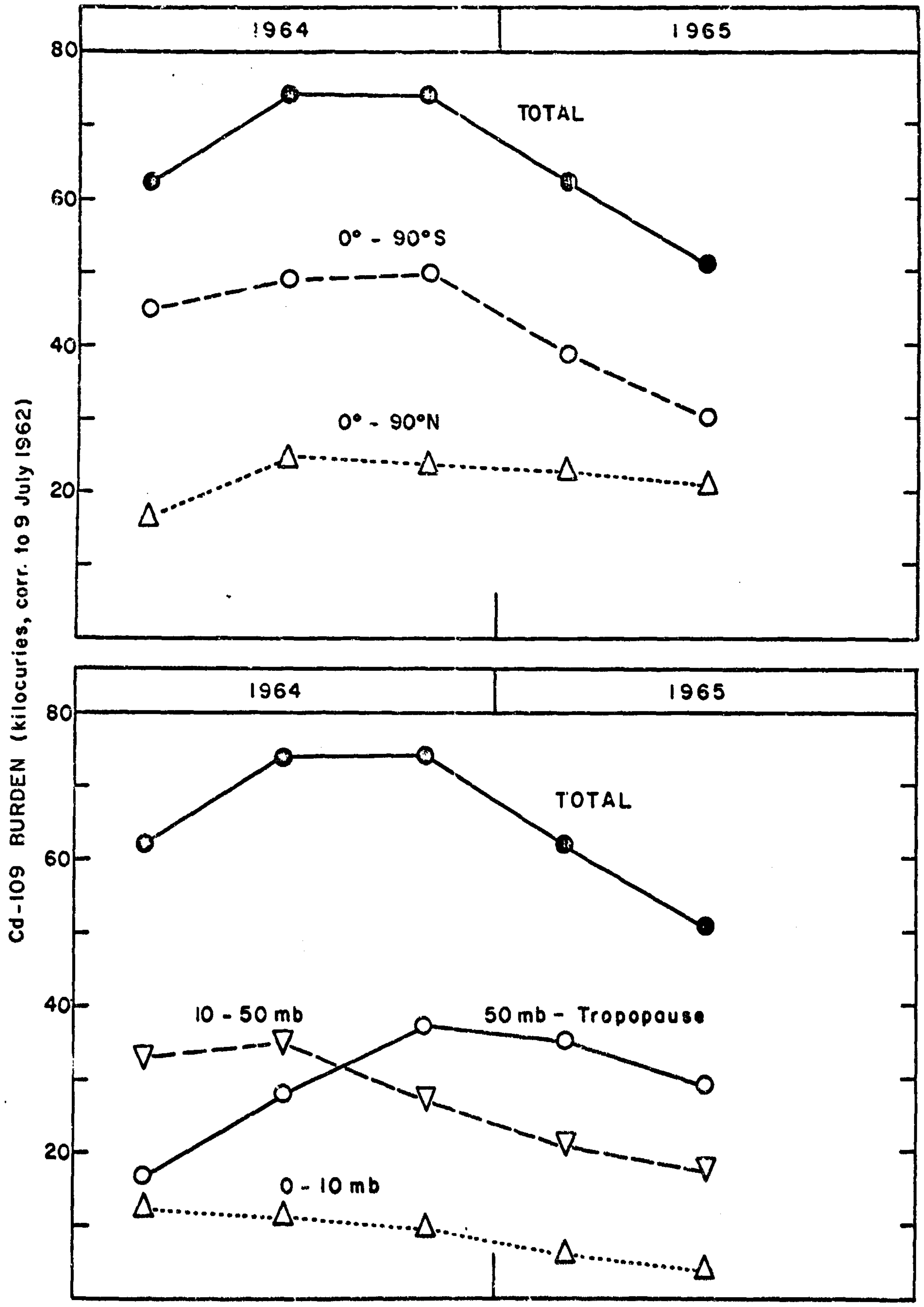


FIGURE 48. STRATOSPHERIC BURDENS OF Cd-109 DURING 1964 AND 1965

The Stratospheric Distribution of Silver-110

In early 1963 some samples collected in the lower polar stratosphere of the Northern Hemisphere were found to contain silver-110. It was hypothesized that this nuclide might have been produced by the Starfish Prime device by means of a $Cd^{110}(n, p)Ag^{110}$ reaction. To check this possibility a series of Stardust samples was analyzed for silver-110.

The results of analyses of samples collected at two points in the Stardust sampling corridor during late 1962 to late 1964 are shown in Figure 49. (In this figure data which represent upper limits of detection are plotted as triangles.) Sufficient data are available for the two points selected, 65 thousand feet at 65°N and at 30°S, to permit comparison of the trends in cadmium-109 and silver-110 activities as a function of time. Cadmium-109 activities at 30°S began increasing in mid-1963, and at 65°N in early 1964 as a result of the influx of debris from Starfish Prime. There is no corresponding increase in the silver-110 concentrations. Moreover, the silver-110 concentrations were an order of magnitude higher in the Northern than in the Southern Hemisphere, while cadmium-109 concentrations reached higher values in the Southern Hemisphere.

The data suggest, then, that silver-110 in the stratosphere is almost certainly a constituent of the radioactive debris from nuclear weapons tests, for it occurs in highest concentration in the stratospheric regions which contain the highest concentrations of such debris. Concentrations of silver-110 show no correlation with those of cadmium-109 in the stratosphere, however, so it is not useful, as is cadmium-109, as a tracer for debris from the Starfish Prime event.

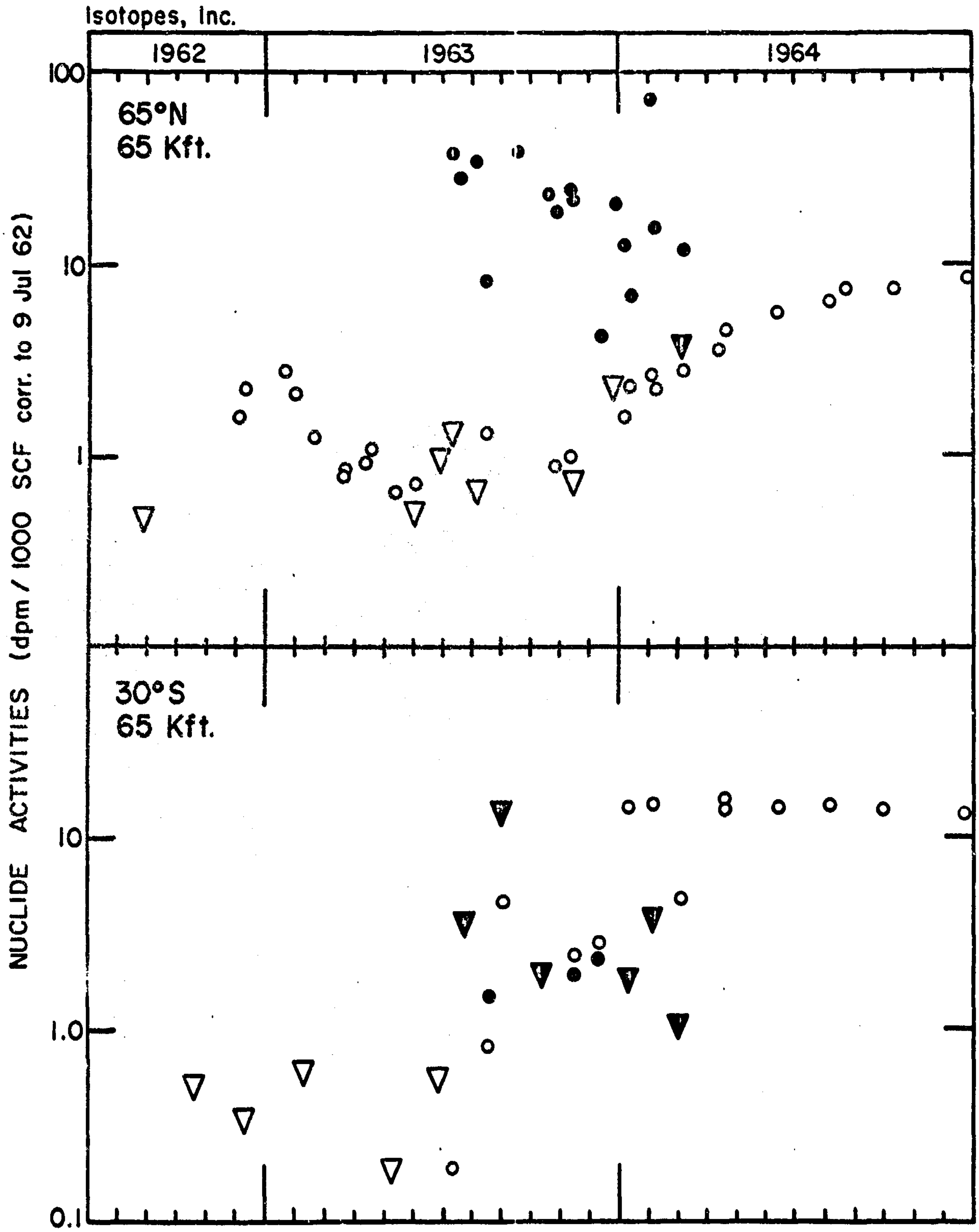


FIGURE 49. TRENDS IN Cd-109 ACTIVITIES (o,∇) AND Ag-110 ACTIVITIES (•,▼) AT 65°N AND 30°S AT 65 Kft.

The Stratospheric Distribution of Carbon-14

Beginning in August, 1963 Stardust sampling missions flown in the region between approximately 70° or 75°N and 15°N have collected compressed air samples as well as filter samples. The collection, processing and analysis of these samples have been described in Chapter 2.

Carbon-14 concentrations in the stratosphere of the Northern Hemisphere have decreased fairly steadily since 1963, as is evident from Figure 50. Bimonthly mean values of carbon-14 concentrations measured at 50 and at 65 thousand feet at 70°N and at 65 thousand feet at 40°N, 25°N and 15°N are plotted in this figure. The increases in carbon-14 concentrations which occurred at 50 thousand feet at 70°N during the winters of 1963-1964 and 1964-1965 may be attributed to an increase in the rate of downward movement of carbon-14 in the polar stratosphere during the winter season, most likely as a result of increased rates of vertical exchange at that time. It is noteworthy that at 65 thousand feet the carbon-14 concentrations in the tropical stratosphere, at 25°N and 15°N, have never reached values comparable to those in the polar stratosphere, at 70°N and 40°N. This behavior differs from that of particulate debris, such as strontium-90 (see Figure 14), which displayed roughly equal concentrations in the tropical and polar stratosphere of the Northern Hemisphere from mid-1963 to mid-1965. It has been hypothesized[6] that the differences in behavior between particulate debris, represented by strontium-90 and manganese-54, and gaseous debris, represented by carbon-14, result from the effects of particle settling in the stratospheric layers above about 65 thousand feet.

The mean distribution of excess carbon-14 in the Stardust sampling corridor has been calculated for each of a series of four month intervals during late 1963 to mid-1965. For calculating concentrations of excess carbon-14, we have assumed that all of the atmosphere contains 74×10^5 atoms of natural carbon-14 per gram of air[1]. The mean distributions of excess carbon-14 during September-December 1963, January-April 1964, and May-August 1964 are shown in Figure 51, and those for

September-December 1964, January-April 1965 and May-August 1965 are shown in Figure 52. There is some evidence that during September-December 1963 there was a layer of maximum concentration at a height of about 60 thousand feet in the polar stratosphere, which sloped upward toward the equator and reached a height of more than 70 thousand feet at 50° to 30°N. During all subsequent periods, however, the highest concentration have been found at the highest altitudes sampled: 65 or 70 thousand feet. The main change in the stratospheric distribution of carbon-14 since the beginning of 1964 has been the gradual decrease of concentrations in the regions above about 40 thousand feet in the northern polar stratosphere and above 50 thousand feet in the northern tropical stratosphere.

The distributions shown in Figures 51 and 52 have been extrapolated into the troposphere, the upper stratosphere and the Southern Hemisphere using results of measurements made at Argonne National Laboratory[1,11], and the stratospheric burdens of excess carbon-14 from nuclear weapons tests have been calculated. The results of these calculations, and estimates of the burdens during the first two thirds of 1963 (based completely on Argonne data) are presented in Table 4 and are plotted in Figure 53. They indicate that the burden of excess carbon-14 in the stratosphere of the Southern Hemisphere changed relatively little between early 1963 and early 1965, but that the burden in the stratosphere of the Northern Hemisphere decreased by about a factor of three between early 1963 and mid-1965. It would appear from Figure 53 that the mean rate of decrease of the total stratospheric burden during 1963 to 1965 corresponded to a stratospheric residence half time of about 18 months, but that the residence time was actually increasing with the passage of time as the fallout rate gradually decreased.

It is of interest to compare changes which occurred in the vertical profiles of carbon-14 activity during 1963-1965 with those which occurred in the vertical profiles of strontium-90 activity. The upper half of Figure 54 contains

mean profiles of excess carbon-14 concentrations at 35°N during September-December 1963, May-August 1964 and January-April 1965. They are based on Stardust measurements of the lower stratosphere, and on Argonne measurements of upper stratospheric balloon samples and of tropospheric aircraft samples[1,11]. The lower half of Figure 54 contains mean profiles of strontium-90 concentrations at about 35°N during the same intervals based on Stardust measurements of the lower stratosphere, and on data from the USAEC high altitude balloon sampling program[2,3,4].

Between late 1963 and mid-1964 both the carbon-14 and strontium-90 concentrations decreased at 35°N at all altitudes above 50 thousand feet. Presumably, most of the decrease at altitudes above the level of the maximum in the profile, 60 to 80 thousand feet, resulted from movement of radioactive debris into the tropical stratosphere or into the southern polar stratosphere. By early 1965 further decreases in carbon-14 concentration had occurred in the layer between 50 and 100 thousand feet, and in strontium-90 concentrations at all altitudes above 50 thousand feet. Again, presumably, the decreases in the region above the level of the maximum may be attributed largely to southward movement of the debris. Decreases at the level of the maximum and at lower levels probably resulted in large part from the loss of the debris from the stratosphere to the troposphere. The failure of the carbon-14 concentrations in the layer between 100 and 110 thousand feet to decrease after mid-1964, while strontium-90 concentrations in that layer were decreasing, is noteworthy. It may provide further evidence of the action of particle fall velocities on the particulate debris in the upper stratosphere. Between late 1963 and early 1965 the strontium-90 concentrations at 105 thousand feet at 35°N decreased by about a factor of 10, while carbon-14 concentrations in the same region decreased only about 30 percent.

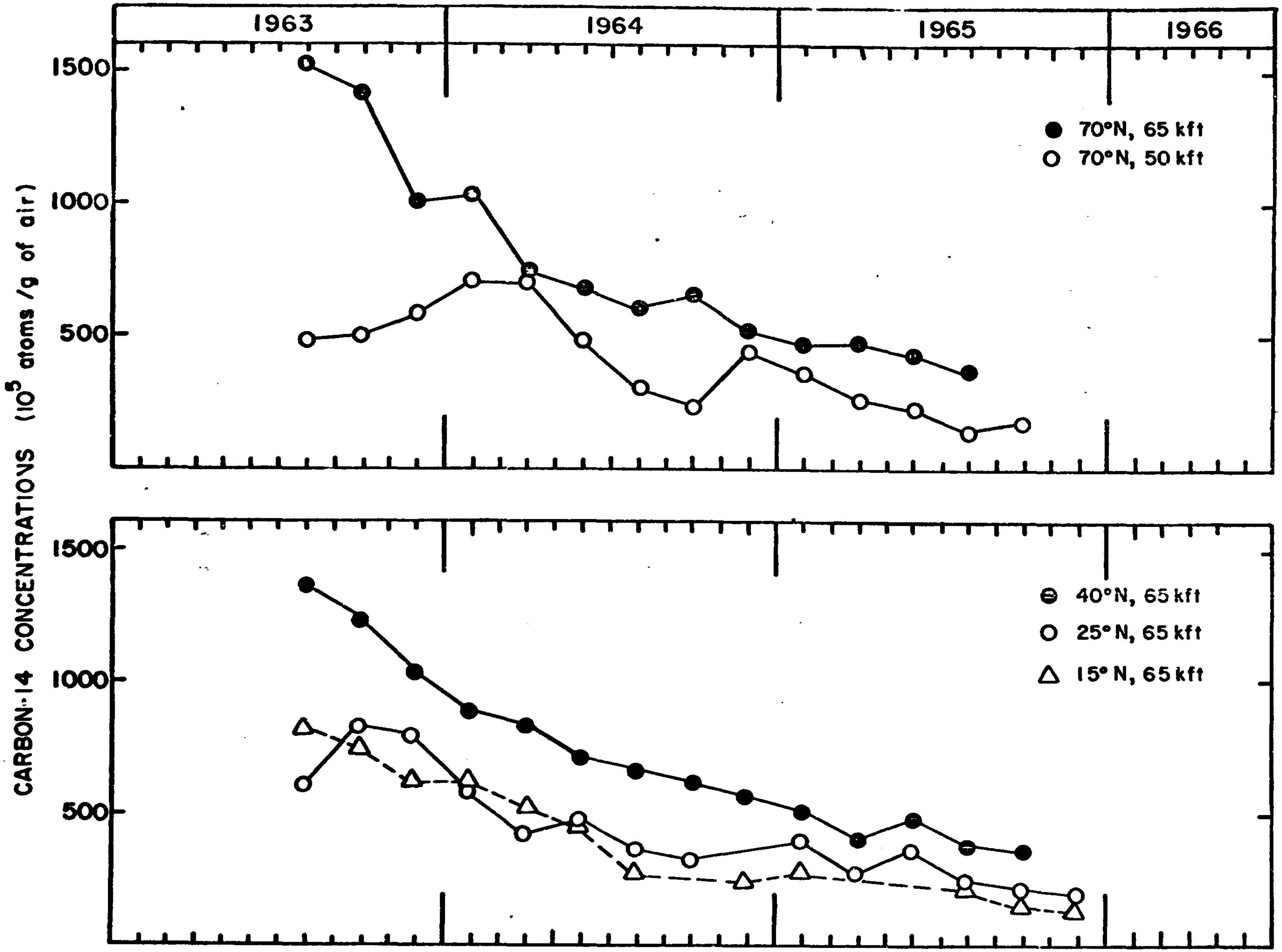


FIGURE 50. TRENDS IN BIMONTHLY MEAN VALUES OF CARBON-14 CONCENTRATIONS

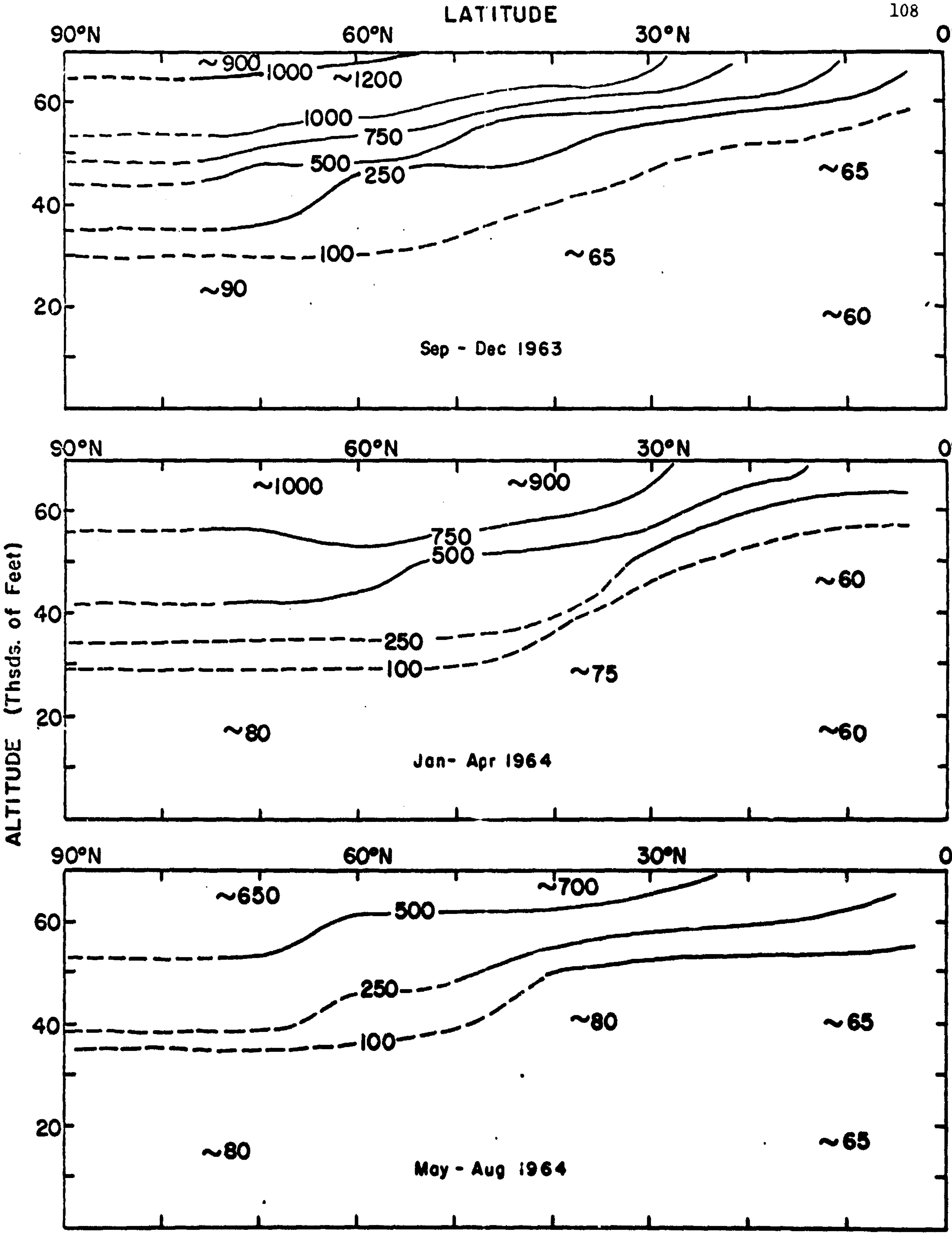


FIGURE 51. MEAN DISTRIBUTION OF EXCESS CARBON-14 (10^5 atoms C^{14}/g air) DURING LATE 1963, EARLY 1964 AND MID-1964

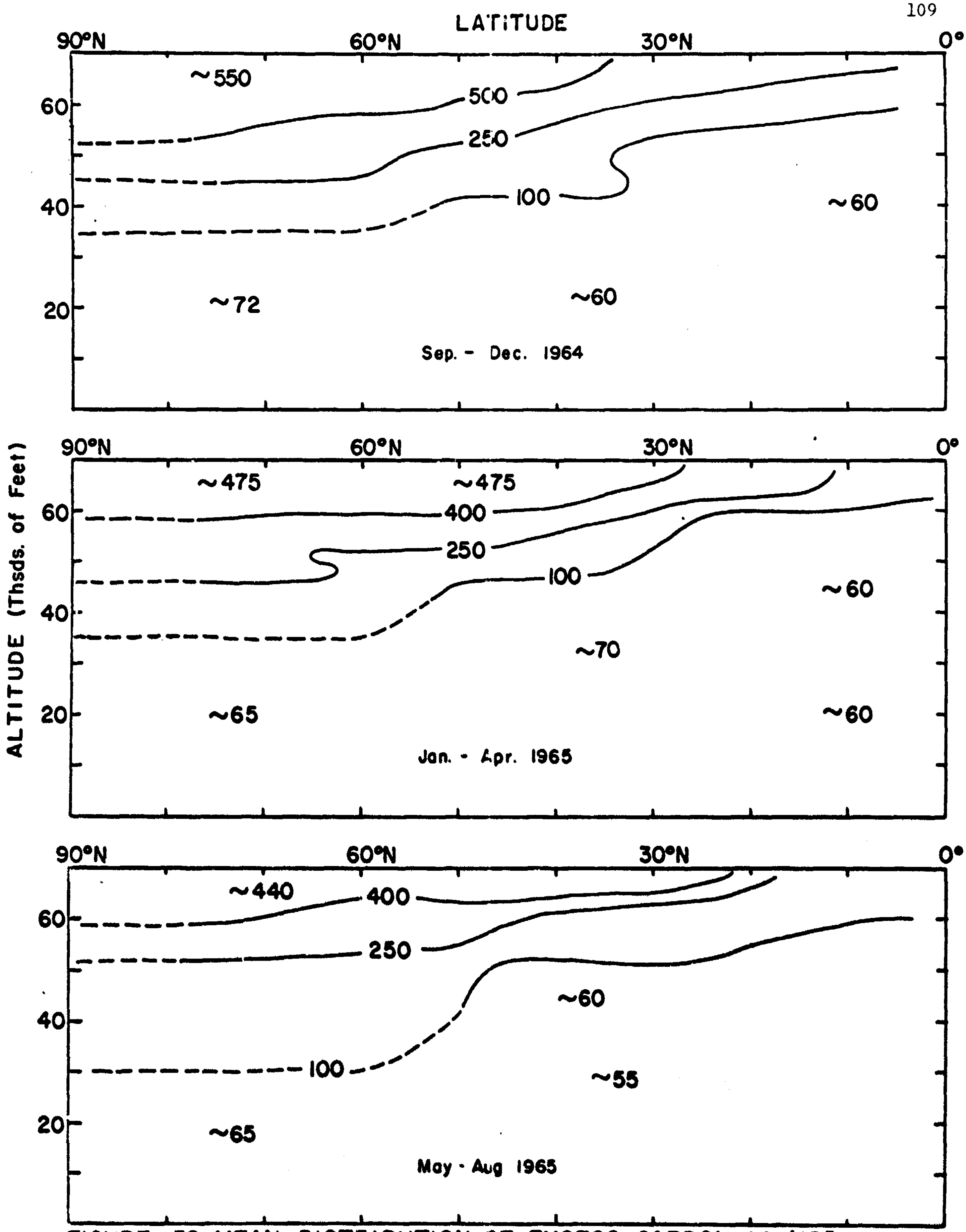


FIGURE 52. MEAN DISTRIBUTION OF EXCESS CARBON-14 (10^5 atoms C^{14} /g air) DURING LATE 1964, EARLY 1965 AND MID 1965

TABLE 4

STRATOSPHERIC BURDENS OF EXCESS CARBON-14 (10^{27} ATOMS)

Time Interval	Northern Hemisphere	Southern Hemisphere	Total Burden
Jan - Apr 1963	30.5 ± 7.0	5.4 ± 3.0	36 ± 8
May - Aug 1963	24.4 ± 6.0	5.3 ± 3.0	30 ± 7
Sep - Dec 1963	20.5 ± 4.0	5.9 ± 3.0	26 ± 5
Jan - Apr 1964	21.3 ± 4.0	5.5 ± 3.0	27 ± 5
May - Aug 1964	14.7 ± 3.5	5.7 ± 2.5	20 ± 4
Sep - Dec 1964	12.6 ± 3.0	5.7 ± 2.5	18 ± 4
Jan - Apr 1965	11.4 ± 3.0	5.7 ± 2.5	17 ± 4
May - Aug 1965	10.2 ± 4.0	-	-

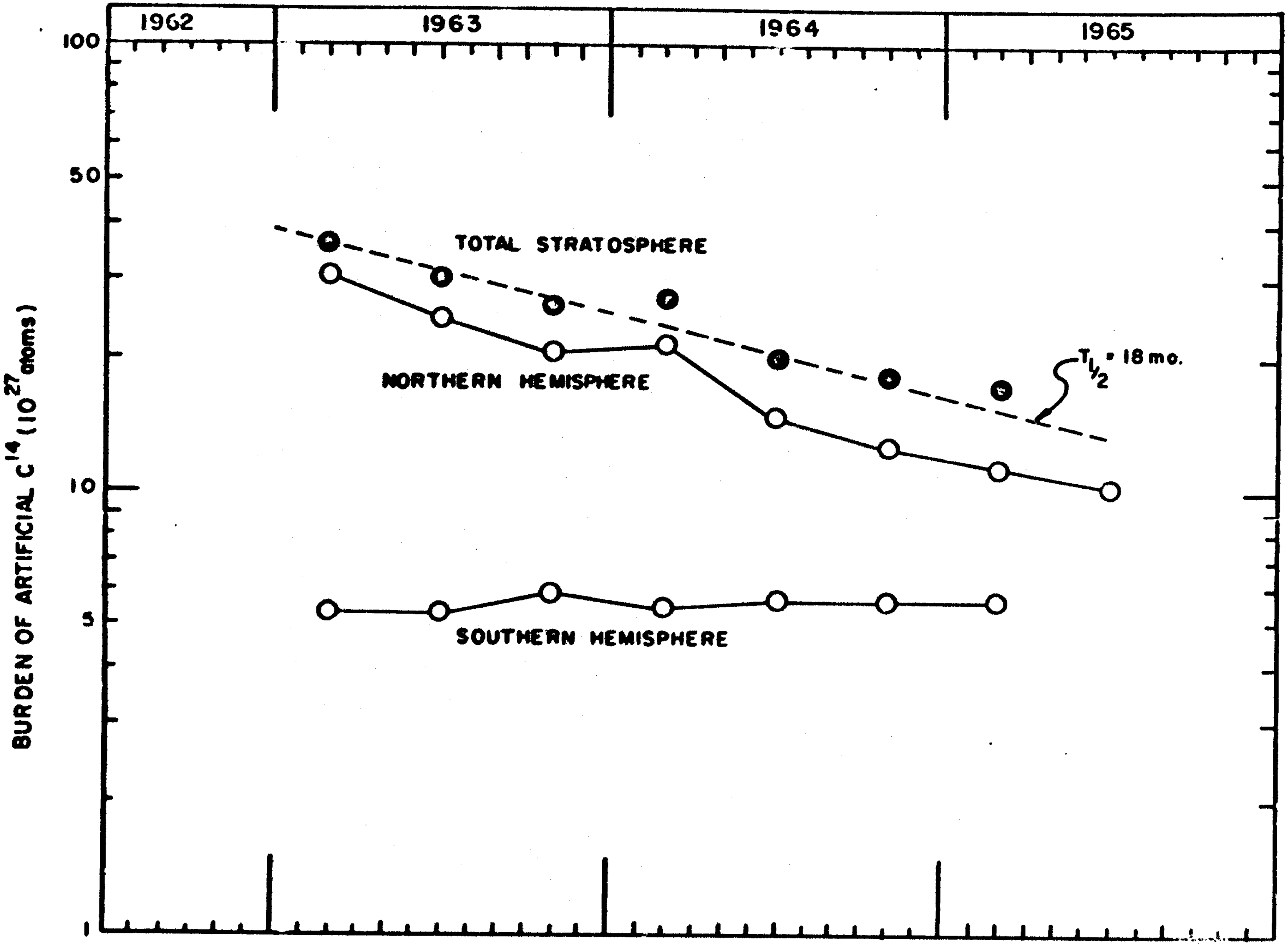


FIGURE 53. STRATOSPHERIC BURDENS OF ARTIFICIAL CARBON-14

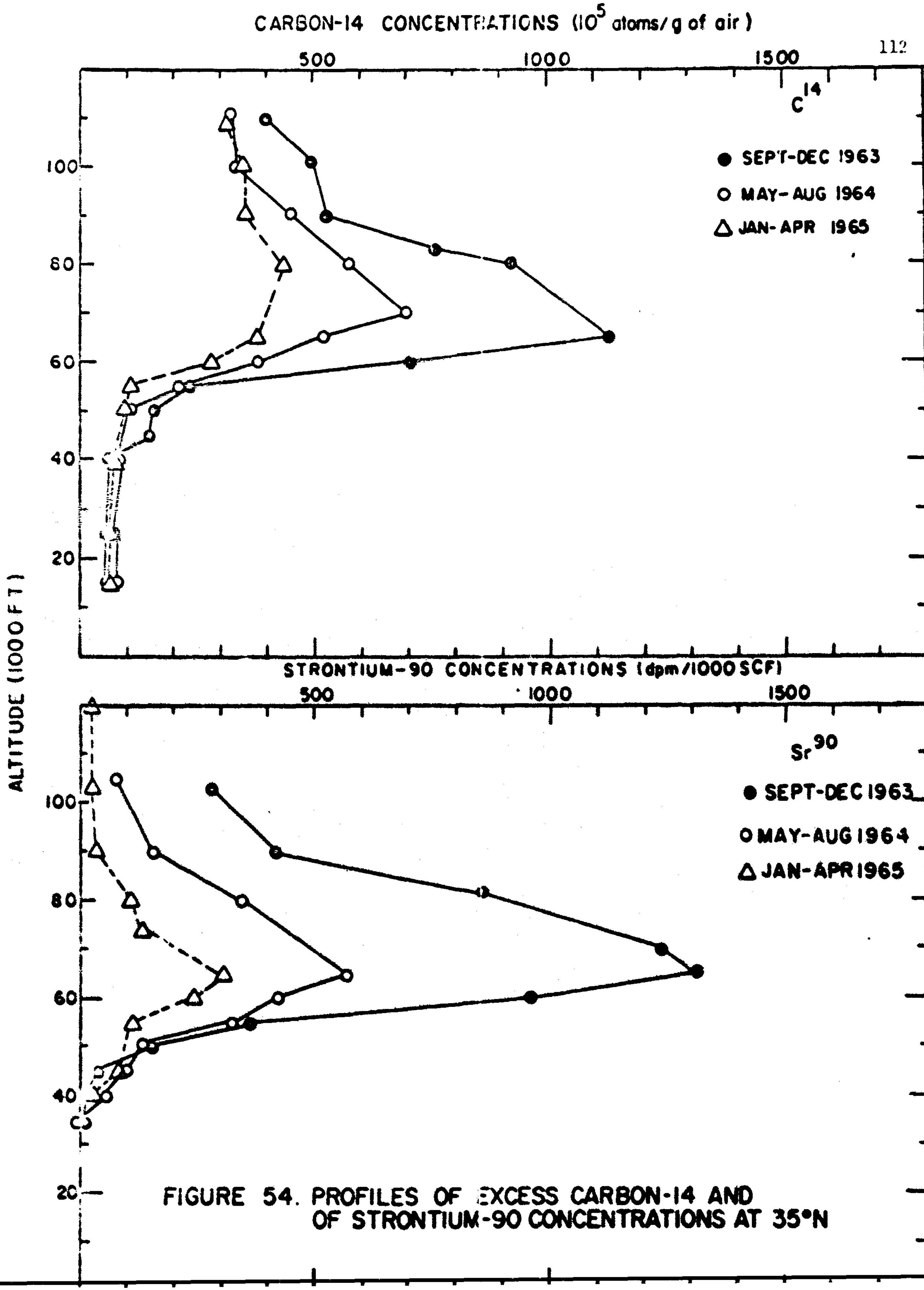


FIGURE 54. PROFILES OF EXCESS CARBON-14 AND OF STRONTIUM-90 CONCENTRATIONS AT 35°N

CHAPTER 5. THE STRATOSPHERIC DISTRIBUTION OF PLUTONIUM-238 FROM THE APRIL, 1964 SNAP-9A BURNUP

Plutonium-238 was injected into the upper stratosphere on 21 April 1964 when a Transit navigational satellite carrying a SNAP-9A power source, which contained 17 kilocuries of plutonium-238, failed to achieve orbit and burned up upon reentry into the atmosphere[12,13,14]. Presumably the debris from the SNAP-9A was distributed initially in the atmospheric layer between 100 and 300 thousand feet altitude, and thus had an initial distribution similar to that of debris from the Orange event in the 1962 Hardtack series of nuclear weapons tests[14]. It will be of great interest, therefore, to monitor the transfer of the SNAP-9A plutonium-238 from the upper to the lower stratosphere, and from the lower stratosphere to the troposphere and to the surface of the earth. The resulting information will be pertinent to studies of fallout rates of debris from nuclear explosions in the upper atmosphere as well as to studies of potential atmospheric contamination resulting from the use of nuclear power sources in space.

The first detection of SNAP-9A plutonium-238 was in filter samples collected by the USAEC high altitude balloon program[15] at 108 thousand feet at 34°S in August, 1964, and in January, 1965 it was detected in the Northern Hemisphere in samples collected at 107 and 109 thousand feet at 31°N. Results of measurements of subsequent balloon samples permitted the tracing of the gradual descent of the SNAP-9A debris to lower levels. The first definite detection of this debris in Stardust samples was in samples collected in May, 1965 at 62 and 65 thousand feet between 38°S and 55°S. The plutonium-238 concentrations in the lower northern polar stratosphere attributable to SNAP-9A debris increased gradually during 1965. By October and November, 1965 half of the plutonium-238 in some samples collected in this region could be attributed to the SNAP-9A source. Stardust sampling was limited

in latitudinal coverage during December 1965, but evidently the influx of SNAP-9A debris into the lower stratosphere of the Northern Hemisphere had greatly accelerated by then, for one sample contained twice as much SNAP-9A plutonium-238 as had any previously collected in that region.

The plutonium-238 concentrations and the Pu^{238}/Pu^{239} activity ratios measured in samples collected during each month between March and December, 1965 are plotted in Figures 55 to 64. In these cross sections a horizontal line is drawn representing the flight track following during the collection of each sample. The plutonium-238 concentrations, in dpm/1000 SCF are given in the upper halves of the figures, and the Pu^{238}/Pu^{239} ratios are given in the lower halves. A diagrammatic representation of the tropopause is given in each figure for general reference. The data are placed in parentheses when it is suspected that an error has been made in the flight data supplied with the sample or during the analysis of the sample.

It appears that Pu^{238}/Pu^{239} ratios between 0.2 and 0.4 are typical of debris from nuclear weapons tests, for results of nearly all analyses of Stardust samples collected before May, 1965 in the Southern Hemisphere and September, 1965 in the Northern Hemisphere fell within that range. No clear sign of SNAP-9A debris was found in samples collected during March, 1965 (Figure 55) or April, 1965 (Figure 56), though it may be significant that of the seven samples collected completely within the Southern Hemisphere during April 1965, the three collected at the highest altitudes displayed slightly higher Pu^{238}/Pu^{239} ratios than did the other four.

During May, 1965 SNAP-9A debris, identified both by relatively high plutonium-238 concentrations and by relatively high Pu^{238}/Pu^{239} ratios, was found in samples collected at 62 and 65 thousand feet at 38°-55°S (Figure 57). Still higher concentrations of SNAP-9A debris were found in this region during June to August, 1965 (Figures 58 to 60). Indeed, the concentrations in the lower southern polar stratosphere generally continued to increase slowly during September to

December, 1965 (Figures 61 to 64). It is evident from the data in Figures 57 to 64 that the SNAP-9A debris was spreading equatorward and downward within the lower southern stratosphere during the final two thirds of 1965.

Apparently a slow influx of SNAP-9A debris into the lower northern polar stratosphere had begun by mid-1965, for beginning in July Pu^{238}/Pu^{239} ratios greater than 0.04 began to appear in samples in that region. Ratios of 0.05 were measured in samples collected there during August and September, 1965 (Figures 60 and 61), and a ratio of 0.06 was found in an October sample (Figure 62), 0.08 in two November samples (Figure 63), and 0.14 in a December sample (Figure 64). Evidently the influx of SNAP-9A debris accelerated with the onset of the winter circulation.

By combining Stardust data with data from the USAEC high altitude balloon sampling program[2,3,4], one may obtain a picture of the downward movement of the SNAP-9A debris within the stratosphere during 1964 and 1965. Results of plutonium-238 measurements performed during these two programs on samples collected at 35°-45°S, at 5°-10°N, at 30°-40°N, and at 65°-70°N are plotted in Figures 65, 66, 67 and 68 respectively.

At 35°-45°S (Figure 65) the plutonium-238 concentrations at 105 and then at 90 thousand feet increased during the late winter and early spring seasons (August-October) of the Southern Hemisphere during 1964 as SNAP-9A debris moved downward from the higher levels where it had been injected in April 1964 (and perhaps as it moved equatorward from higher latitudes where most of the downward movement may actually have occurred. Little additional downward movement of the SNAP-9A debris occurred at 35°-45°S during October 1964 to March 1965 (the summer season of the Southern Hemisphere), but during April to August 1965 (the autumn and the winter seasons) the debris moved downward again, reaching the level of the tropical tropopause in detectable quantities. This downward motion of the debris resulted in about a tenfold decrease in the plutonium-238 concentrations at 105 thousand feet. After August, 1965 any additional downward movement of the SNAP-9A debris at 35°-45°S was slow, as the winter circulation gave way to the spring and

summer circulation of the stratosphere.

At 5°-10°N (Figure 66), in the tropical stratosphere SNAP-9A debris was present at 105 thousand feet by the beginning of 1965, but had not reached 65 thousand feet by November, 1965. As yet, no samples collected at 5°-10°N have shown plutonium-238 concentrations as high as those found in either the southern or the northern polar stratosphere.

In the northern polar stratosphere (Figures 67 and 68) SNAP-9A plutonium-238 reached 90 thousand feet, coming from the region above 105 thousand feet, during the winter of 1964-1965, and reached 80 thousand feet during the spring and summer of 1965. It was not until December 1965, after the winter circulation of the polar stratosphere had again been established in the Northern Hemisphere, that it reached 65 thousand feet at 30°-40°N (and presumably also at 65°-70°N), where it was sampled by Stardust aircraft.

The value of the Pu^{238}/Pu^{239} activity ratio was approximately 0.032 in Stardust samples collected in the Southern Hemisphere before May, 1965 and in the Northern Hemisphere before November, 1965. Using this value, then, as the Pu^{238}/Pu^{239} ratio in debris from nuclear weapons tests, we have calculated the weapons component of plutonium-238 in each Stardust sample, based on the measured plutonium-239 concentration in the sample. We have subtracted this weapons component from the total plutonium-238 concentration of each sample to determine the SNAP-9A component.

The concentrations of SNAP-9A plutonium-238 in samples collected at 40°S and in samples collected at 25°S have been plotted in time - altitude sections in Figure 69 and concentration isolines have been drawn through the data points. At 40°S SNAP-9A plutonium-238 reached 65 thousand feet during May, 1965 and reached the vicinity of the polar tropopause at 40 thousand feet in August, 1965. At 25°S the SNAP-9A plutonium-238 probably reached 65 thousand feet during June, 1965, and reached the vicinity of the tropical tropopause at 53 thousand feet by September

1965. It is evident that the mechanisms of atmospheric transport which cause the movement of radioactive debris from the upper into the lower stratosphere are capable of transferring significant quantities of the debris through virtually the entire thickness of the lower stratosphere over the course of a few months. Such rapid transfer is probably more readily explained in terms of eddy diffusion than in terms of mean advective transport within the stratosphere.

After August, 1965 changes in the distribution of SNAP-9A plutonium-238 at 40°S occurred only slowly. Presumably this reflects a decrease in the rate of vertical exchange with the onset of the summer circulation in the southern stratosphere. The decrease in concentrations in the layer between 40 and 55 thousand feet at 40°S after October, 1965 probably resulted from the poleward migration of the jet-stream to latitudes higher than 40° during the spring months, with the resultant replacement of the low polar tropopause by the high tropical tropopause at 40°S.

The mean distributions of total plutonium-238 and of SNAP-9A plutonium-238 in the Stardust sampling corridor during September to December, 1965 have been estimated, and with the help of data from the AEC high altitude balloon program they have been extrapolated into the upper atmosphere. These distributions are plotted in Figure 70. Based on these distributions the stratospheric burden of SNAP-9A plutonium-238 during September to December, 1965 has been calculated. The results are summarized in Table 5.

The calculated total stratospheric burden of SNAP-9A plutonium-238 is 7.5 kilocuries, which is less than half of the 17 kilocuries presumably injected in April, 1964, by the SNAP-9A power source[12,13,14]. Only 2.4 kilocuries were found in the lower stratosphere (below 40 millibars), where Stardust aircraft could sample it. Our estimation of the burden in the upper stratosphere, based on incomplete results of late 1965 balloo sampling, could easily be in error by a factor of two, and this could explain most of the discrepancy between the amount injected and the

calculated burden. Part of the remaining discrepancy might be attributable to an overestimate of the collection efficiencies of the balloon samplers at the higher altitudes. If these efficiencies are actually less than 100 percent the calculated burdens of SNAP-9A plutonium-238, most of which was still present at high altitudes in late 1965, would be too low.

Two samples collected between 17°S and about 35°S, one during May and one during June, 1965 appeared to contain SNAP-9A plutonium-238. These results seem anomalous, however, for samples collected farther south at the same altitude and at higher altitudes at the same latitude did not contain SNAP-9A debris. These two samples, SQ-7251 and SQ-7264, were reanalyzed, as SQ-7565 and SQ-7566 respectively, to confirm these potentially significant results. In the original analyses one half of each filter was used, and in the reanalyses one quarter of each was used. Data from the analyses of these four samples are contained in Table 6. The reanalyses failed to confirm the presence of SNAP-9A plutonium-238 in the samples.

The failure of the reanalyses to find SNAP-9A debris probably means that the original analyses were in error, but there is another possibility. If the SNAP-9A debris which entered the lower stratosphere during the first half of 1965 was carried by particles large enough to contain between 10^8 to 10^9 atoms of plutonium-238, only one or two such particles would have to be included in a sample to give the level of plutonium-238 activity displayed by samples SQ-7251 and SQ-7264. The sizes of the hypothetical spherical particles of $Pu^{238}O_2$ required to contain all of the SNAP-9A plutonium-238 in each of the two samples, SQ-7251 and SQ-7264, have been calculated. The calculation, which is shown in Table 7, indicates radii of 0.15 and 0.12 micron for these hypothetical spherical particles. If the SNAP-9A plutonium-238 were actually present as aggregates of smaller sphericle particles, and the aggregates consisted of 50 percent open spaces and 50 percent particles of millimicron size, the radii of the spherical aggregates would be 26 percent larger than is calculated in Table 4.

No evidence is currently available to confirm the possible correctness of the original analyses and of the reanalyses of the two anomalous samples, but the alpha spectra obtained in both sets of analyses appear to be of good quality. Data are included in Table 6 for sample SQ-7564, collected at 38°-55°S one month before the first definite interception there of SNAP-9A debris, and for sample SX-7476, collected at approximately the same location after comparatively high concentrations of SNAP-9A debris had arrived. The alpha spectra for these two samples are plotted in Figure 71. Peaks are shown which are attributable to plutonium-239, plutonium-238 and plutonium-236 (which is added to the samples before analysis to permit calculation of radiochemical yields). The addition of SNAP-9A plutonium-238 to plutonium from weapons debris causes a readily discernible increase in the ratio of the area under the 5.5 Mev plutonium-238 peak to the area under the 5.1 Mev plutonium-239 peak. Alpha spectra for samples SQ-7251 and SQ-7565 and for samples SQ-7264 and SQ-7566 (all with the 5.8 Mev plutonium-236 peaks deleted) are compared in Figure 72. All spectra are of reasonably good quality, suggesting that perhaps the analyses are indeed correct, and that failure of the plutonium-238 results to agree on duplicate samples results from inhomogeneous distribution of SNAP-9A debris on the filters. This could result if the particles carrying the debris had radii in the range of 0.12 to 0.15 micron. Some small studies which may provide evidence for or against this possibility are being pursued, including additional reanalyses of these and other filters and autoradiography of a filter containing relatively high concentrations of SNAP-9A plutonium-238. Unless additional evidence is found to support the results for samples SQ-7251 and SQ-7264, however, we will assume that they were in error.

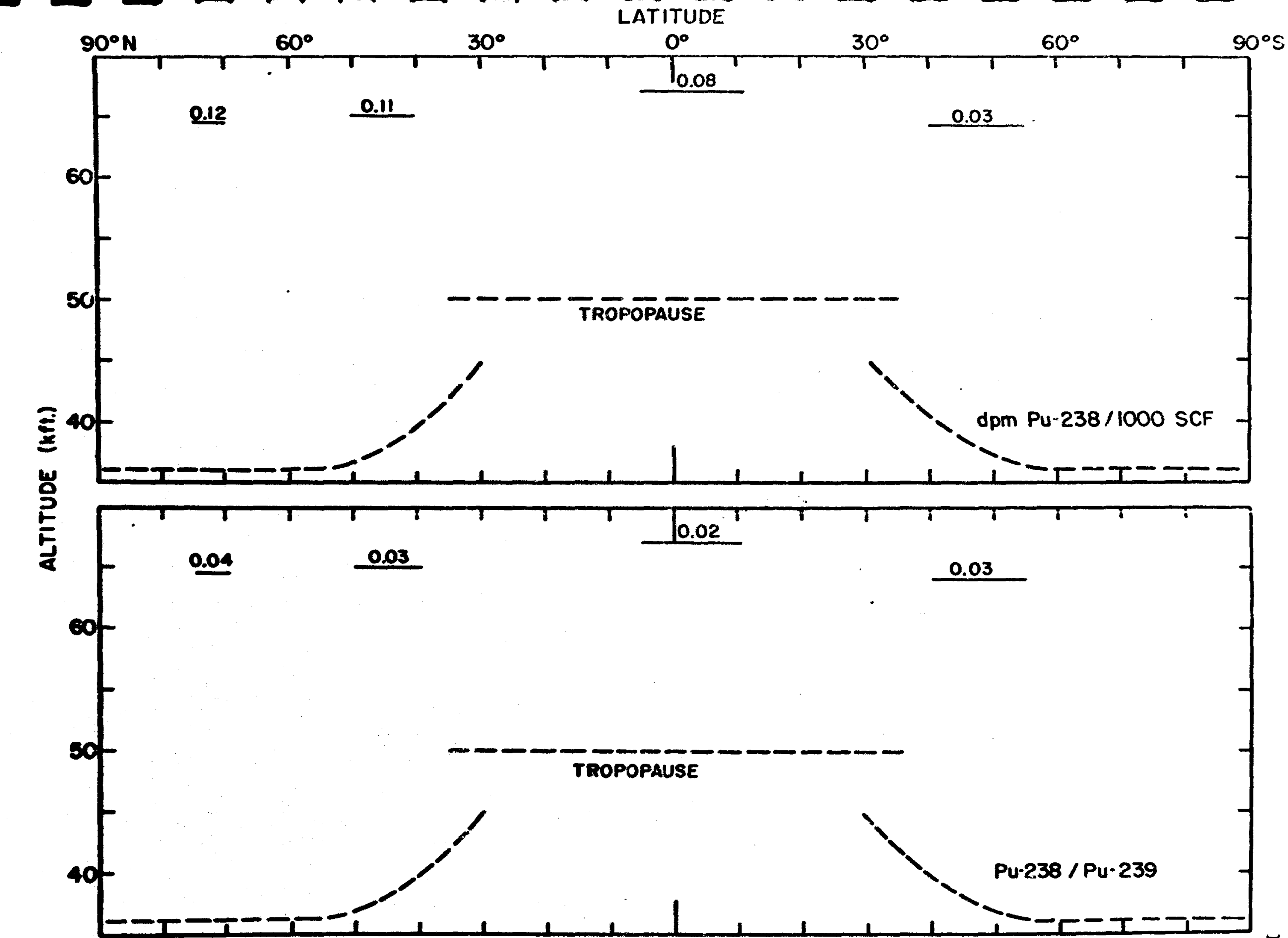


FIGURE 55. DISTRIBUTION OF Pu-238 AND OF THE ACTIVITY RATIO Pu-238/Pu-239 DURING MARCH 1965

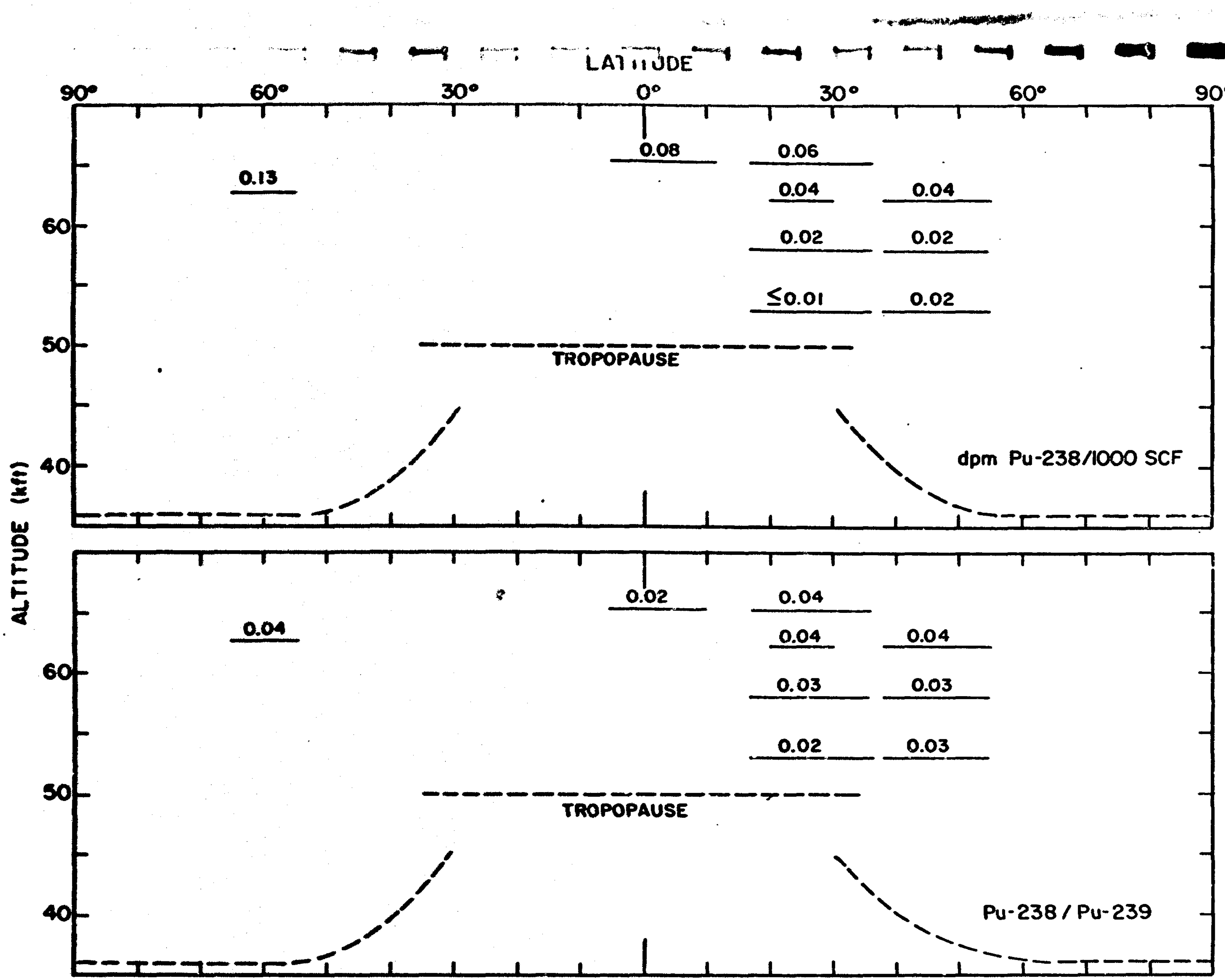


FIGURE 56. DISTRIBUTION OF Pu-238 AND OF THE ACTIVITY RATIO Pu-238/Pu-239 DURING APRIL 1965

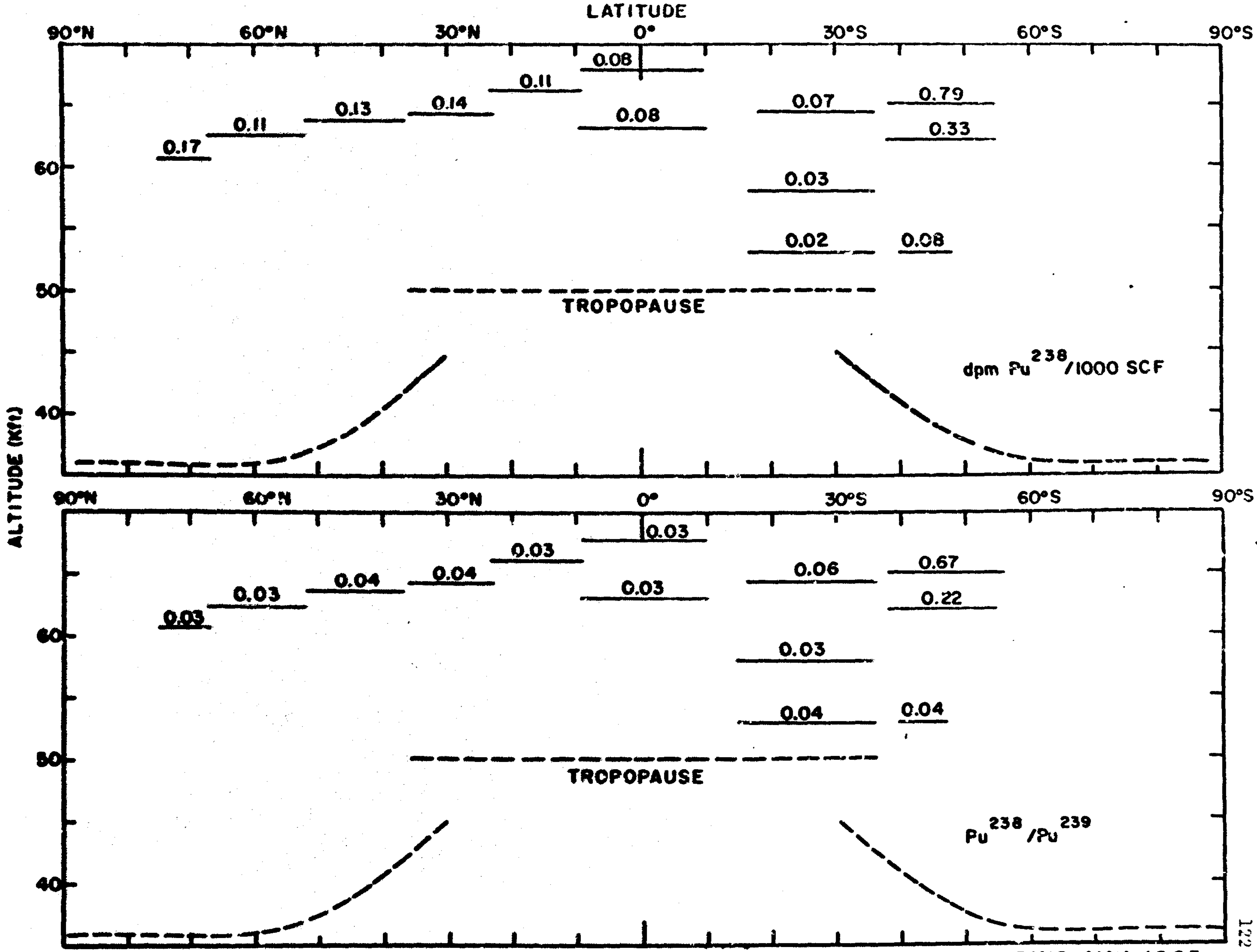


FIGURE 57. DISTRIBUTION OF Pu-238 AND OF THE ACTIVITY RATIO Pu-238/Pu-239 DURING MAY 1965

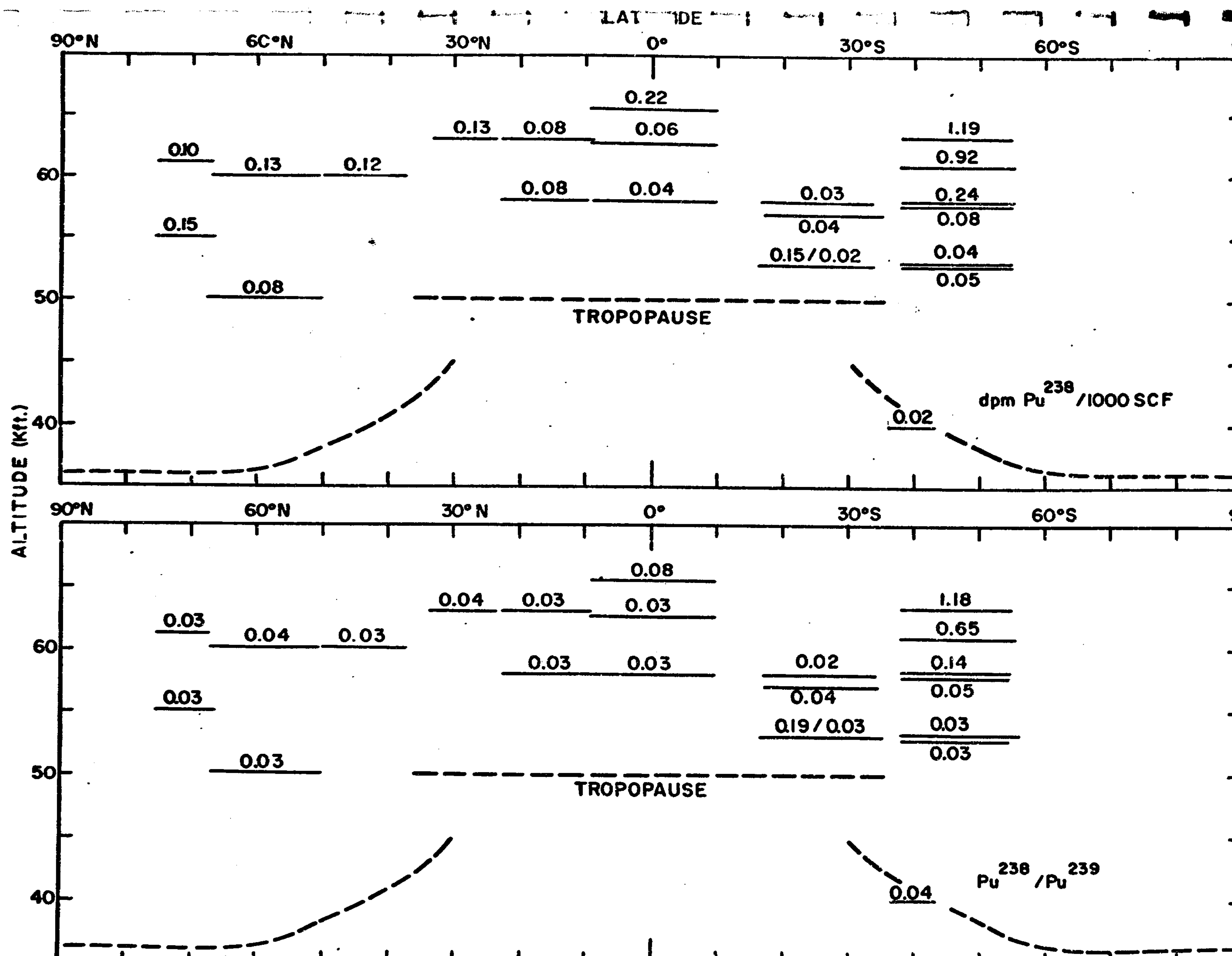


FIGURE 58. DISTRIBUTION OF Pu-238 AND OF THE ACTIVITY RATIO Pu-238/Pu-239 DURING JUNE 1965

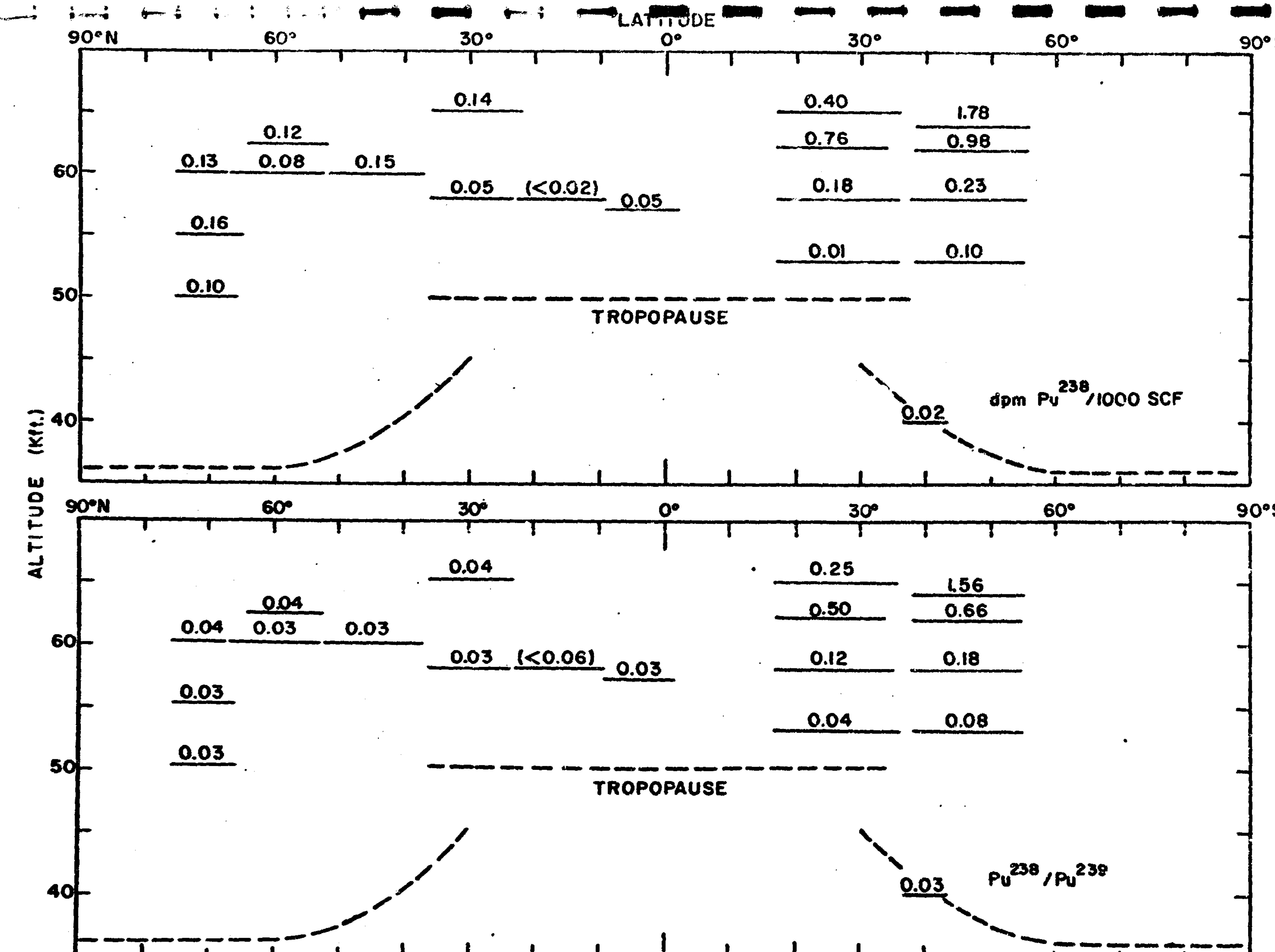


FIGURE 59. DISTRIBUTION OF Pu-238 AND OF THE ACTIVITY RATIO Pu-238/Pu-239 DURING JULY 1965

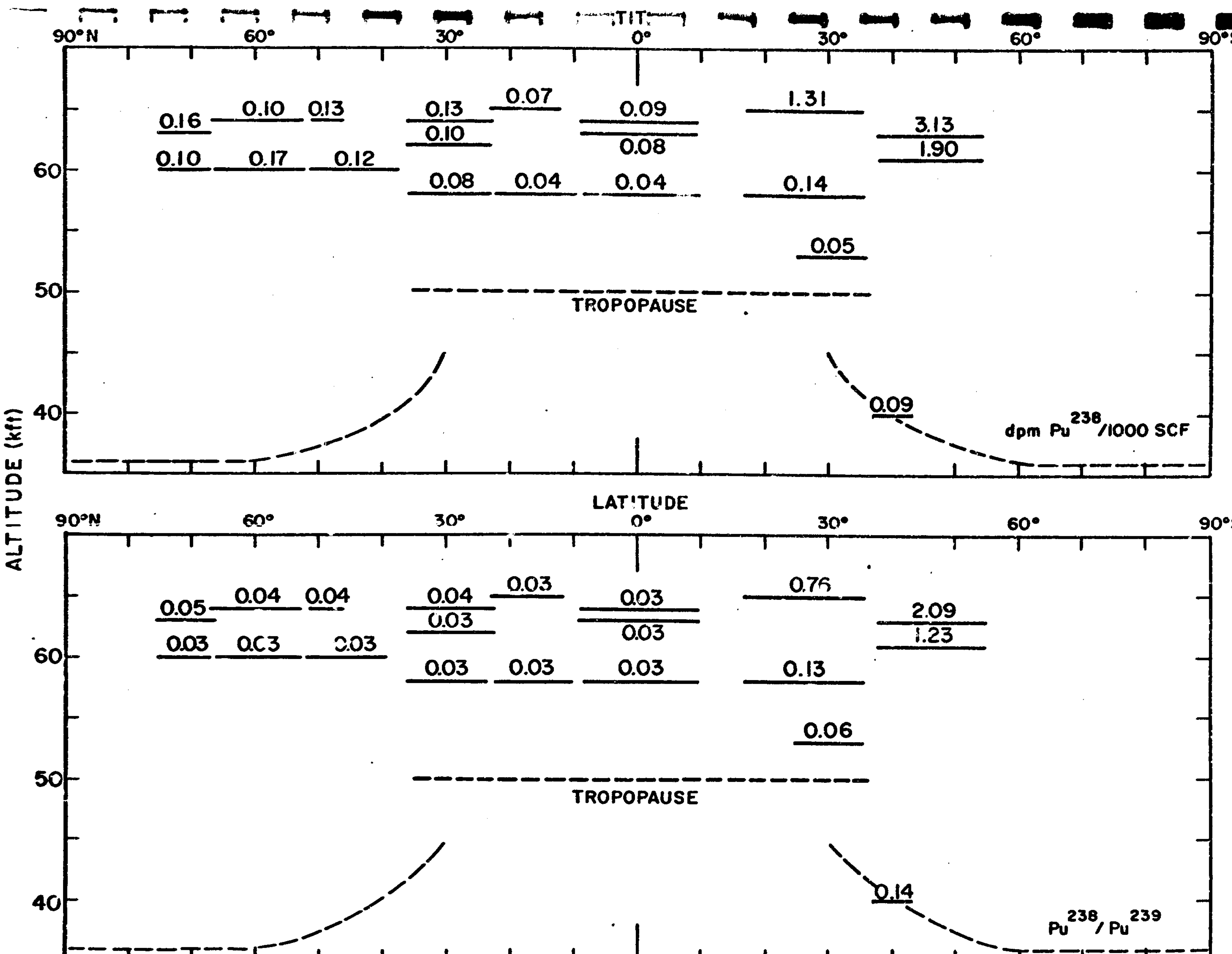


FIGURE 60. DISTRIBUTION OF Pu-238 AND OF THE ACTIVITY RATIO Pu^{238}/Pu^{239} DURING AUGUST 1965

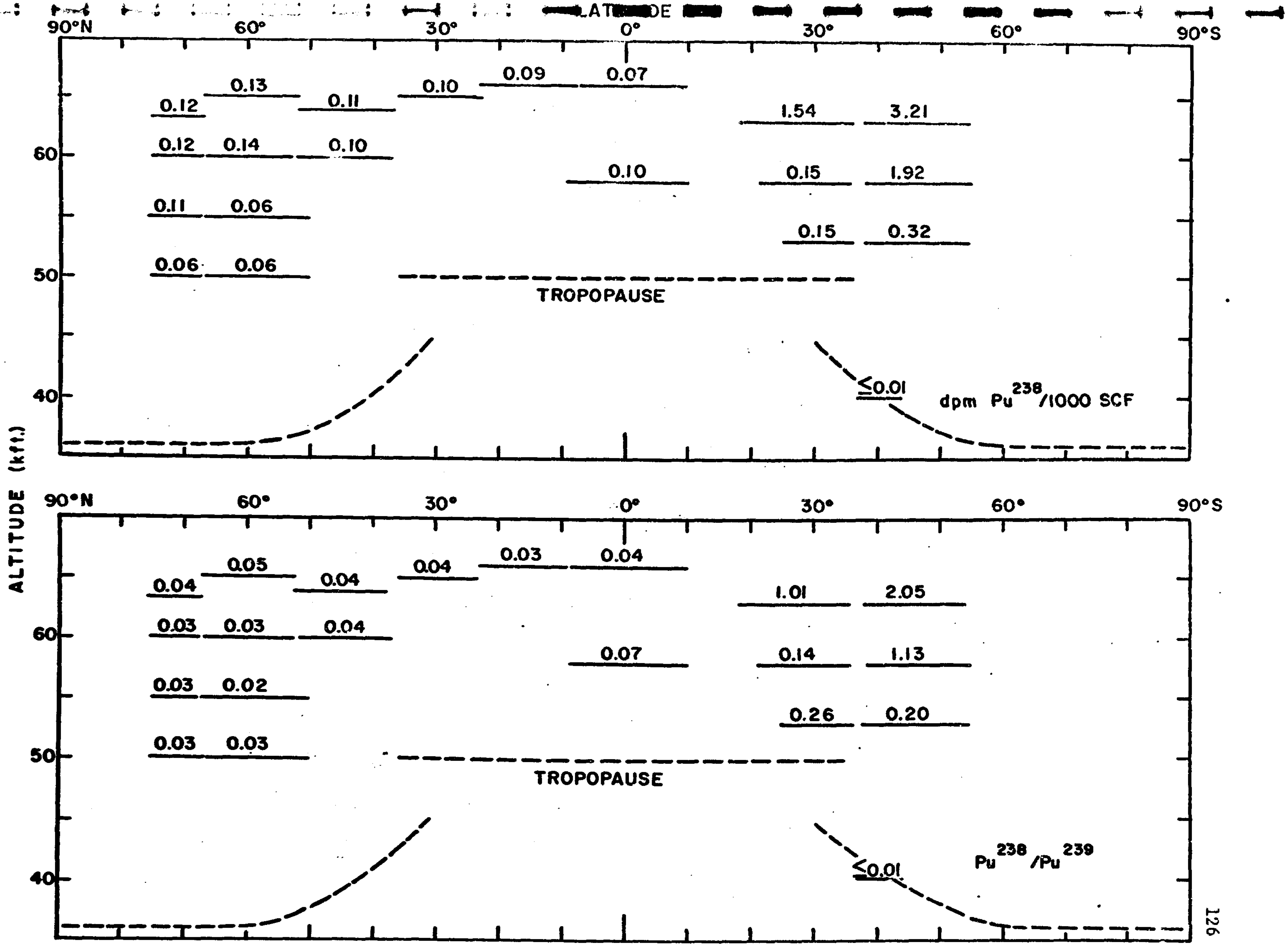


FIGURE 61. DISTRIBUTION OF Pu-238 AND OF THE ACTIVITY RATIO Pu-238/Pu-239 DURING SEPT. 1965.

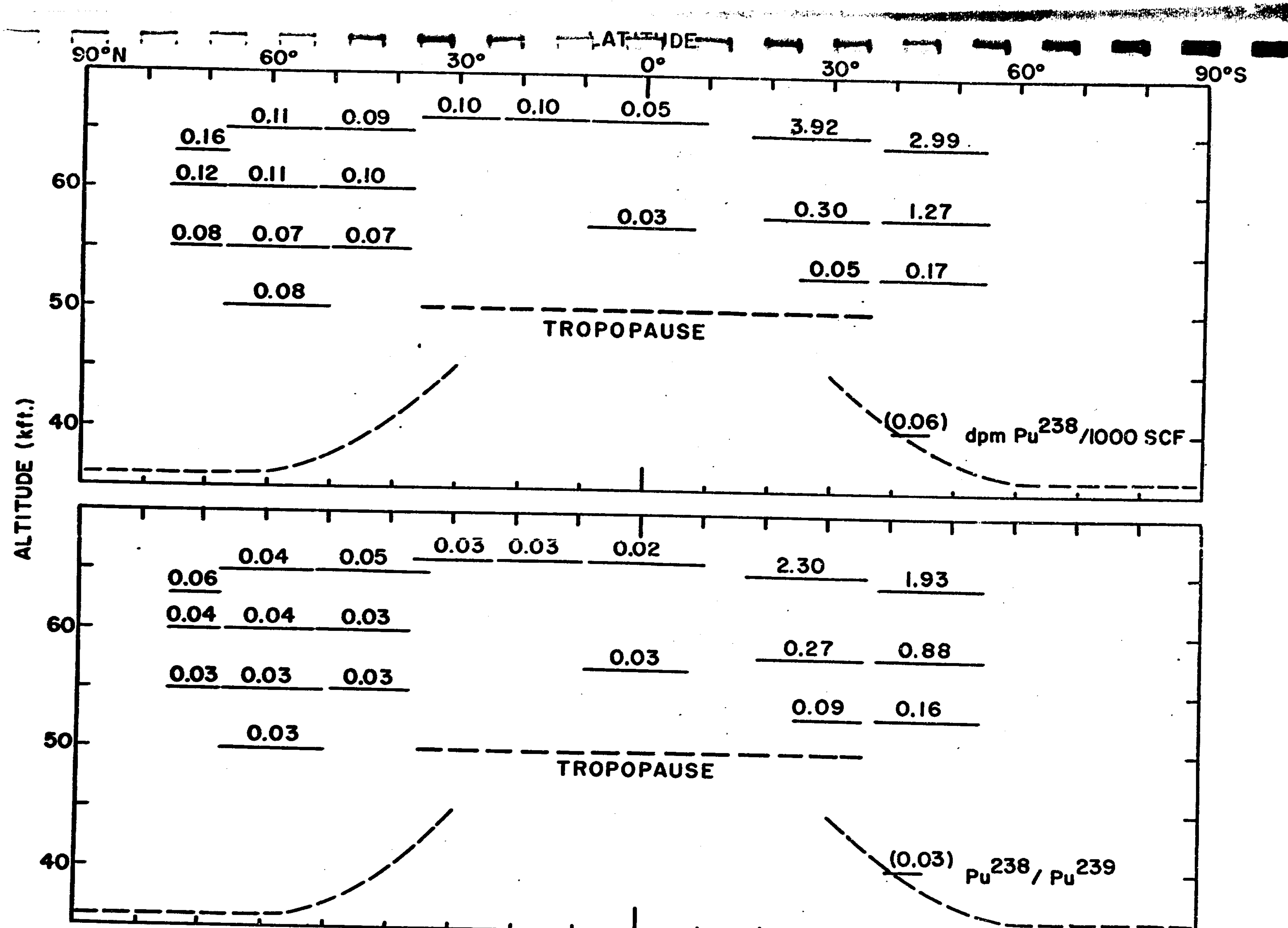


FIGURE 62. DISTRIBUTION OF Pu-238 AND OF THE ACTIVITY RATIO Pu-238/Pu-239 DURING OCT. 1965

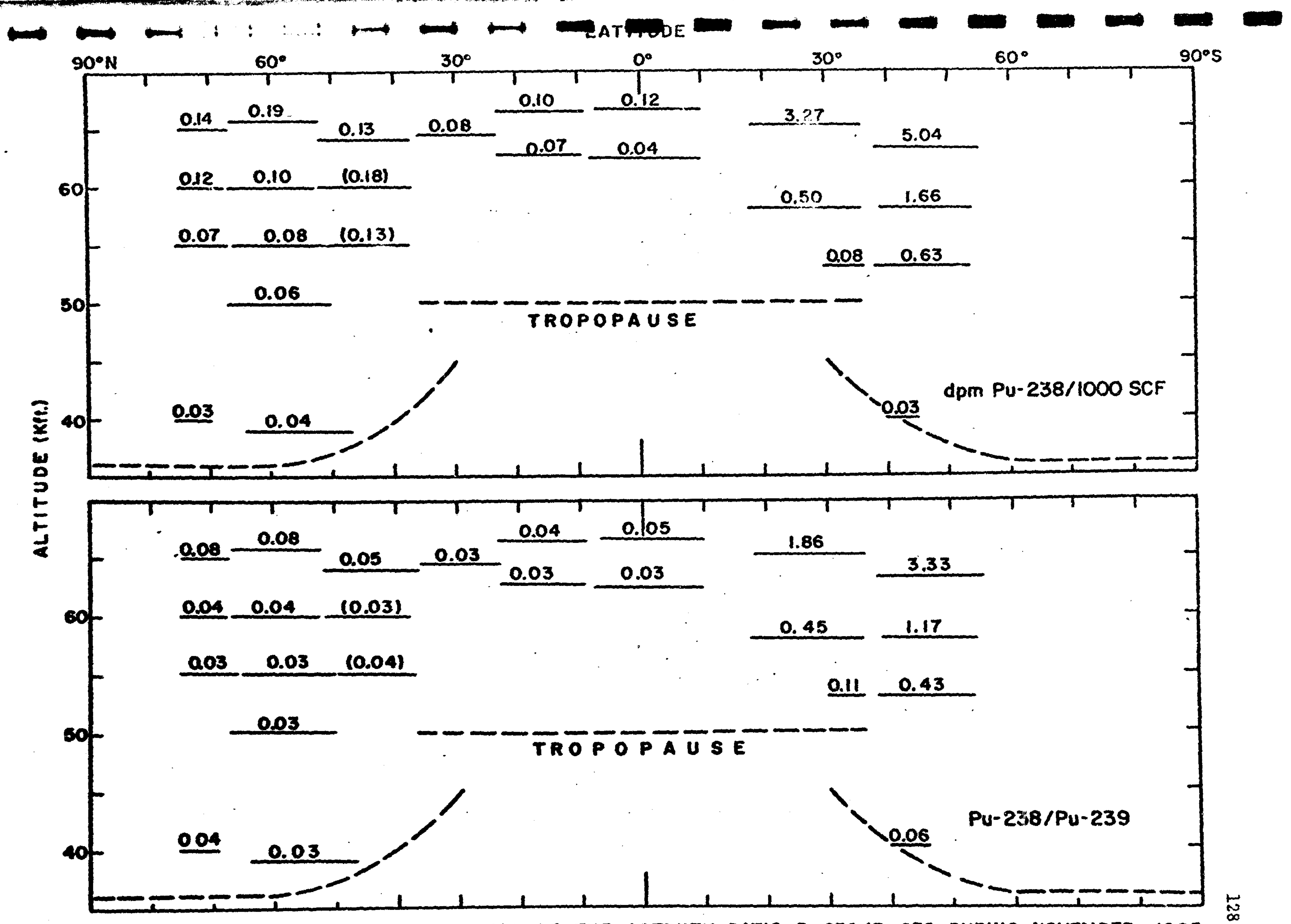


FIGURE 63. DISTRIBUTION OF Pu-238 AND OF THE ACTIVITY RATIO Pu-238/Pu-239 DURING NOVEMBER 1965.

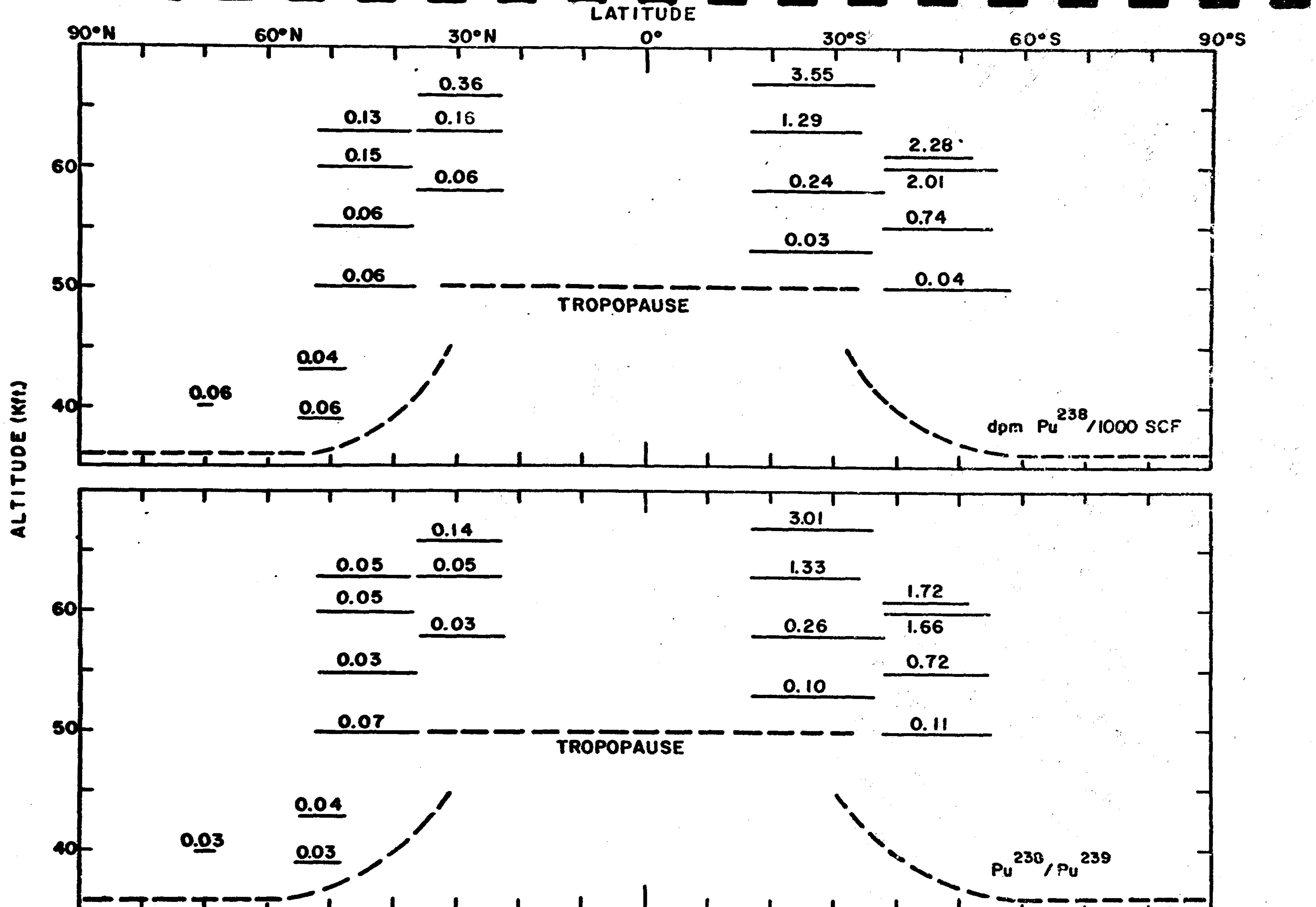


FIGURE 64. DISTRIBUTION OF Pu^{238} AND OF THE ACTIVITY RATIO Pu^{238}/Pu^{239} DURING DECEMBER 1965.

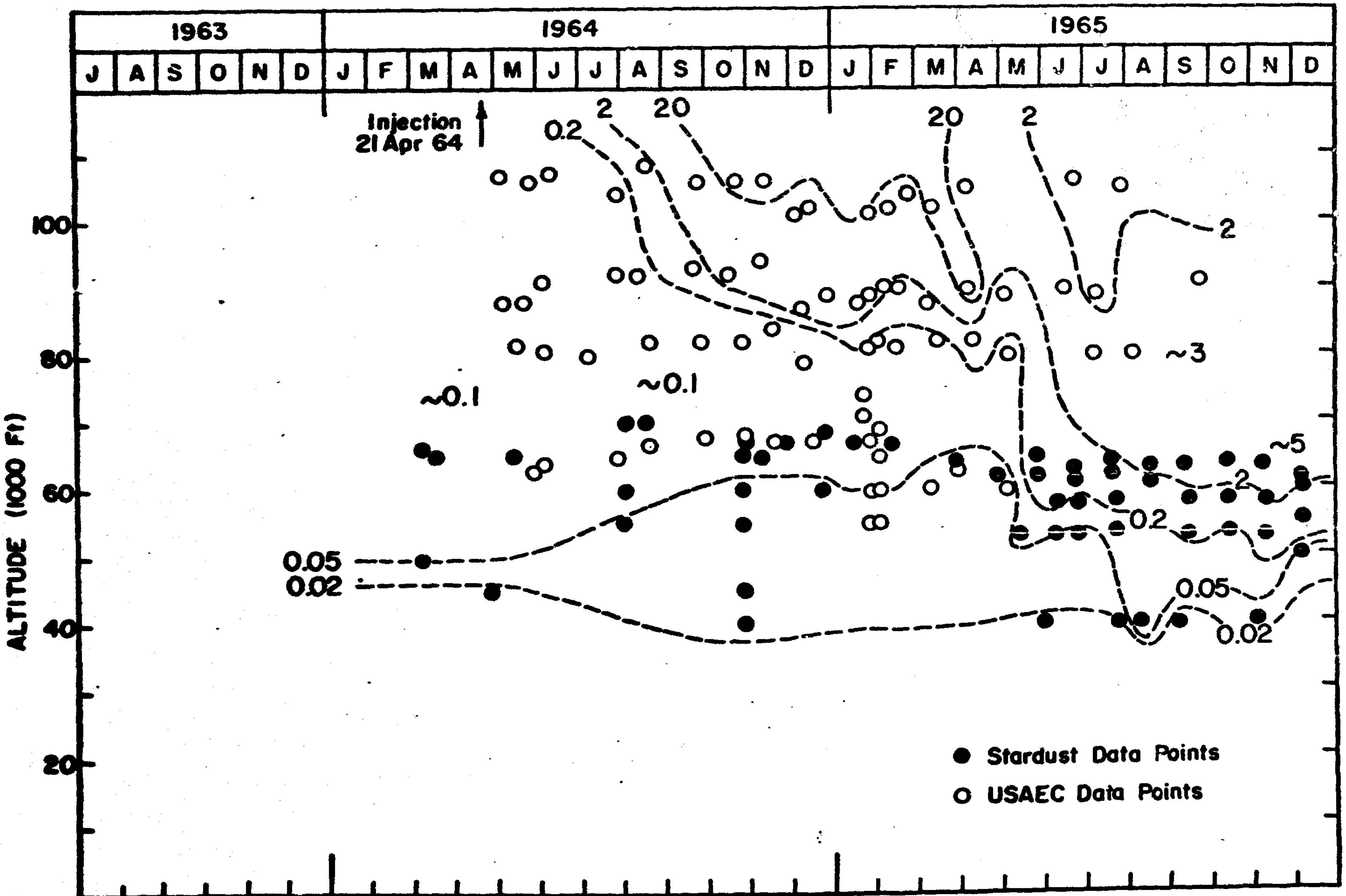


FIGURE 65. PLUTONIUM-238 ACTIVITIES (dpm Pu-238/1000 SCF) AT 35°-45°S DURING 1964 - 1965

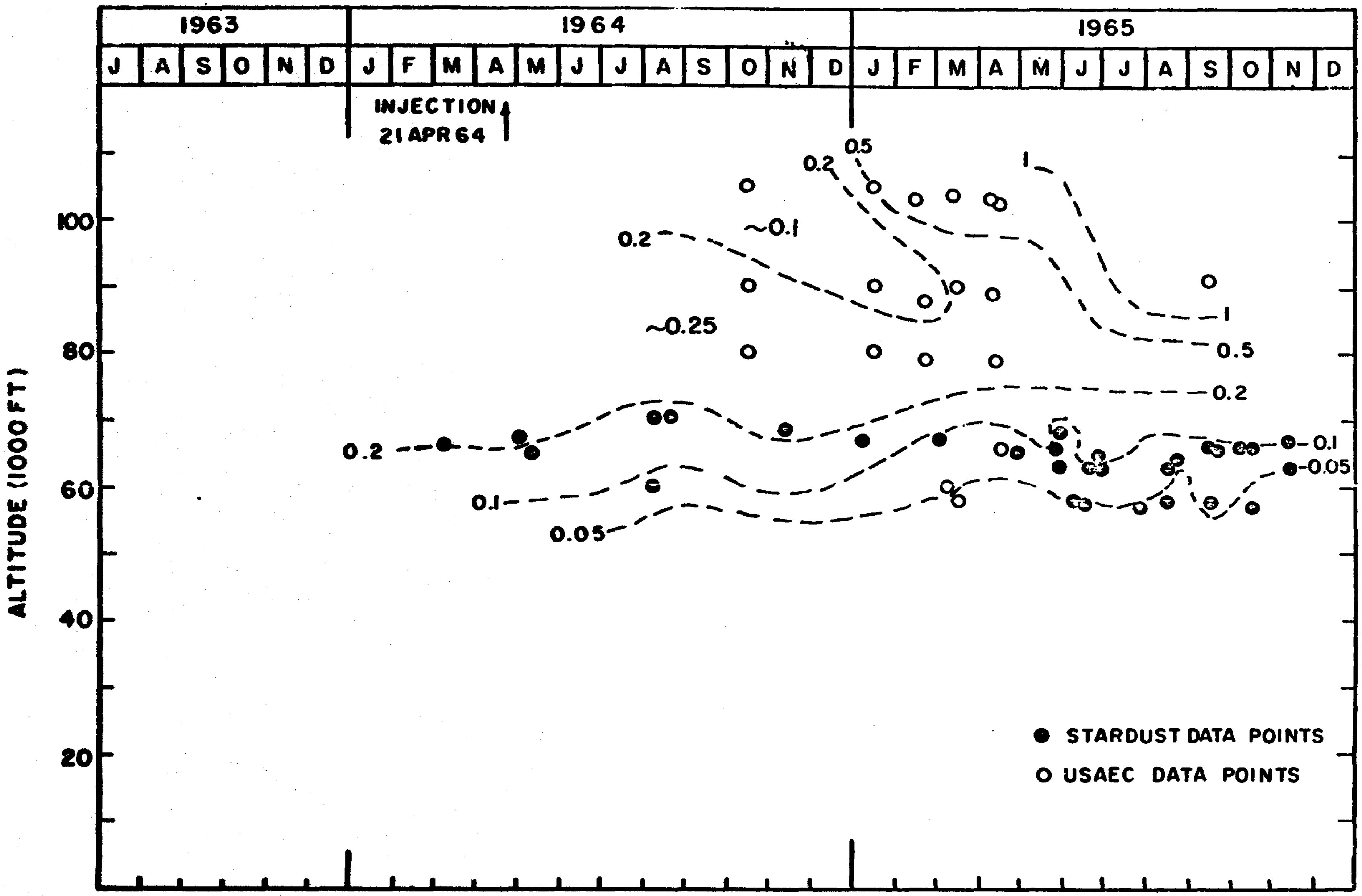


FIGURE 66. PLUTONIUM-238 ACTIVITIES (dpm Pu^{238}/1000 SCF) AT 5°-10°N DURING 1964-1965

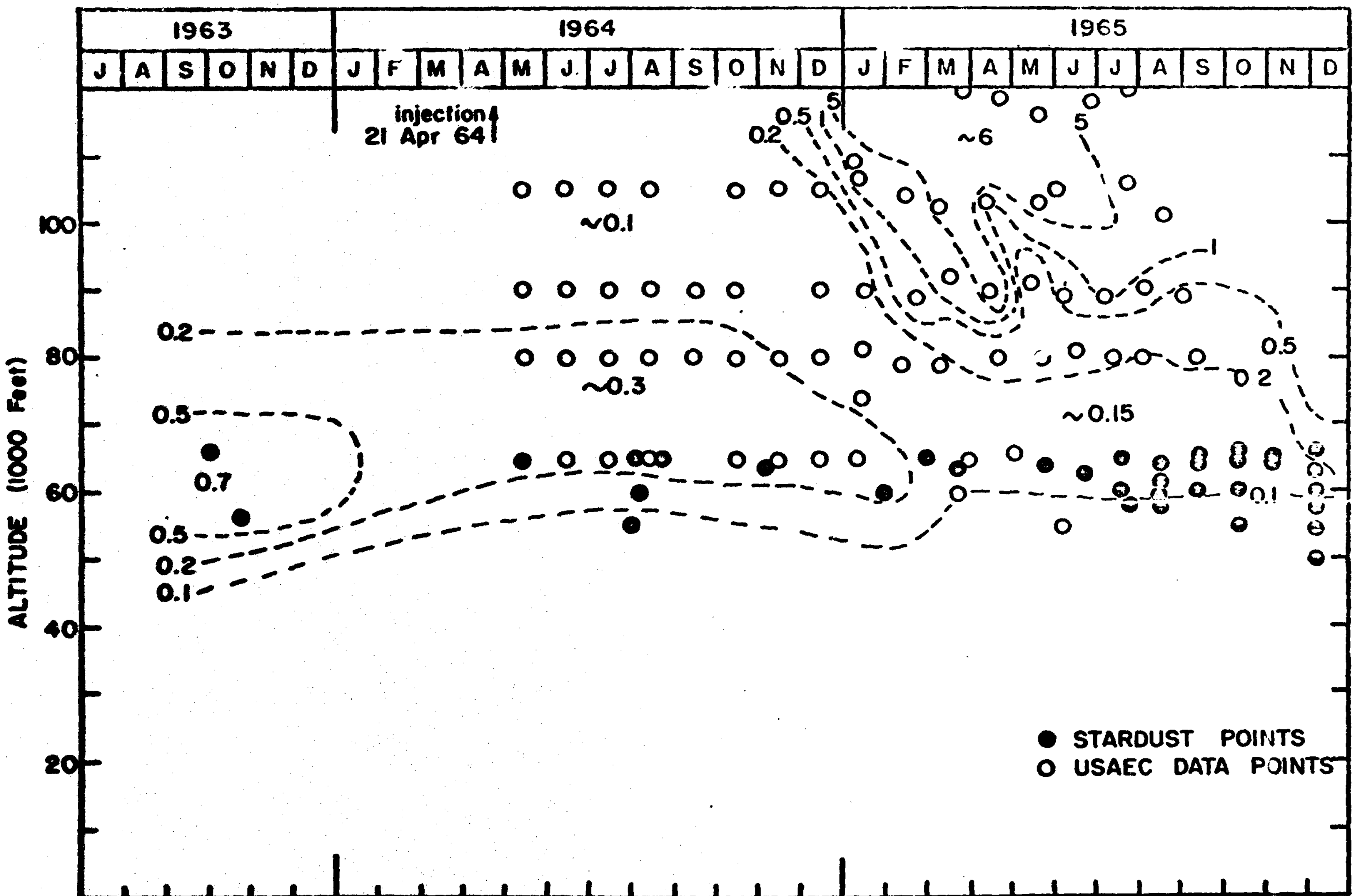


FIGURE 67. PLUTONIUM-238 ACTIVITIES (dpm Pu-238/1000 SCF) AT 30°-40°N DURING 1963 - 1965

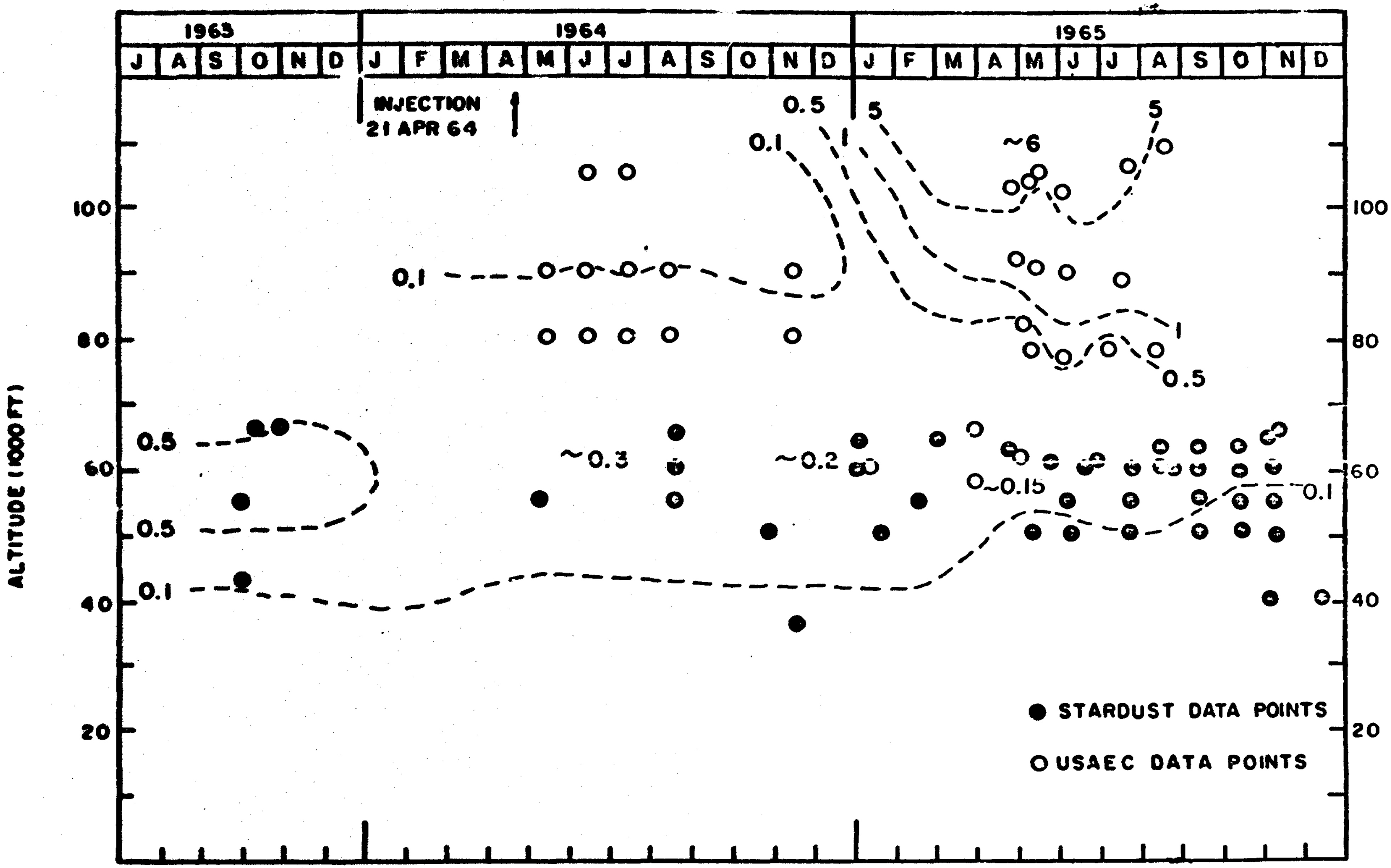


FIGURE 68. PLUTONIUM-238 ACTIVITIES (dpm PU^{238}/1000 SCF) AT 65°–70°N DURING 1963–1965

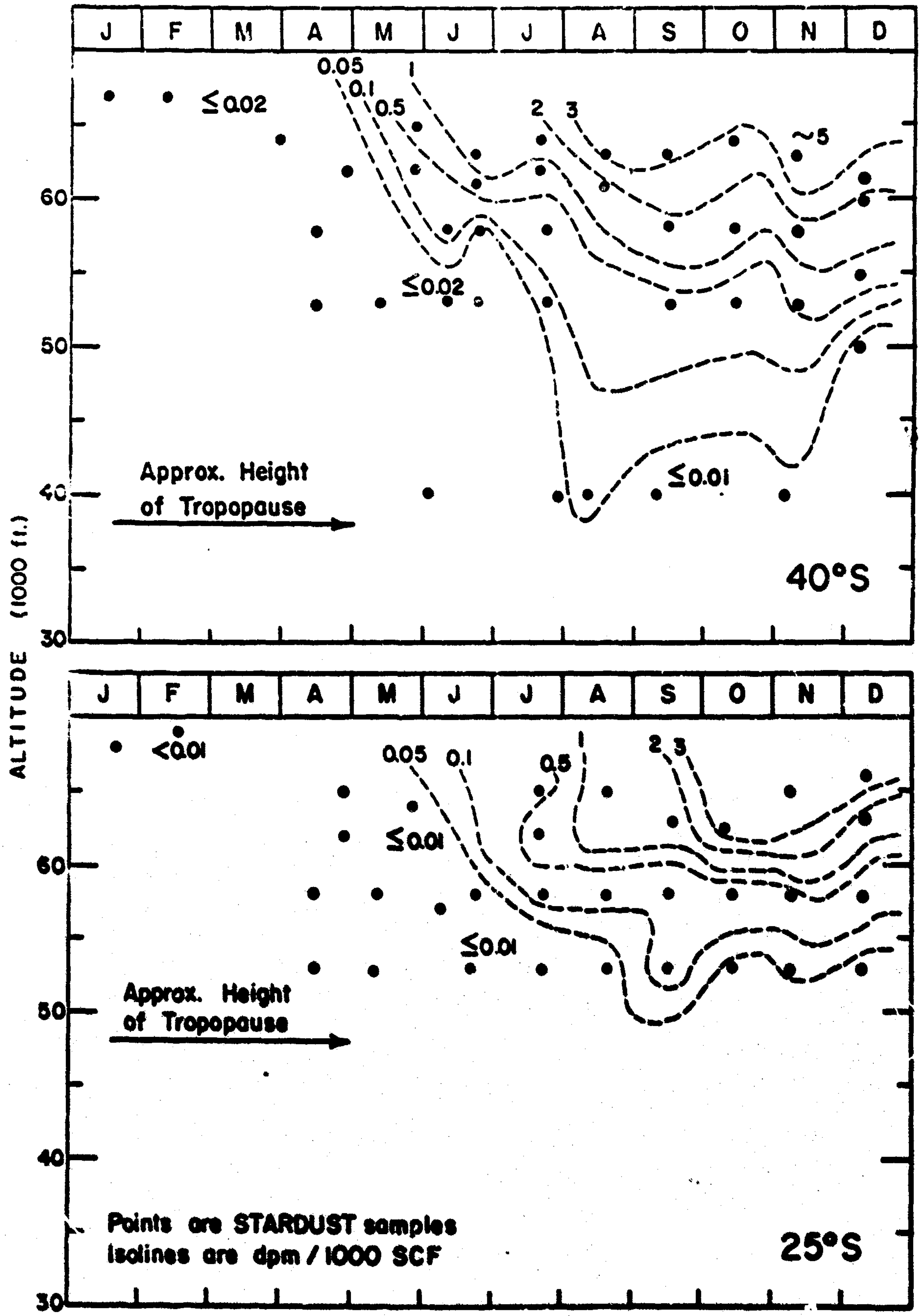


FIGURE 69. TIME-ALTITUDE DISTRIBUTION OF SNAP-9A Pu-238 AT 40°S AND 25°S DURING 1965

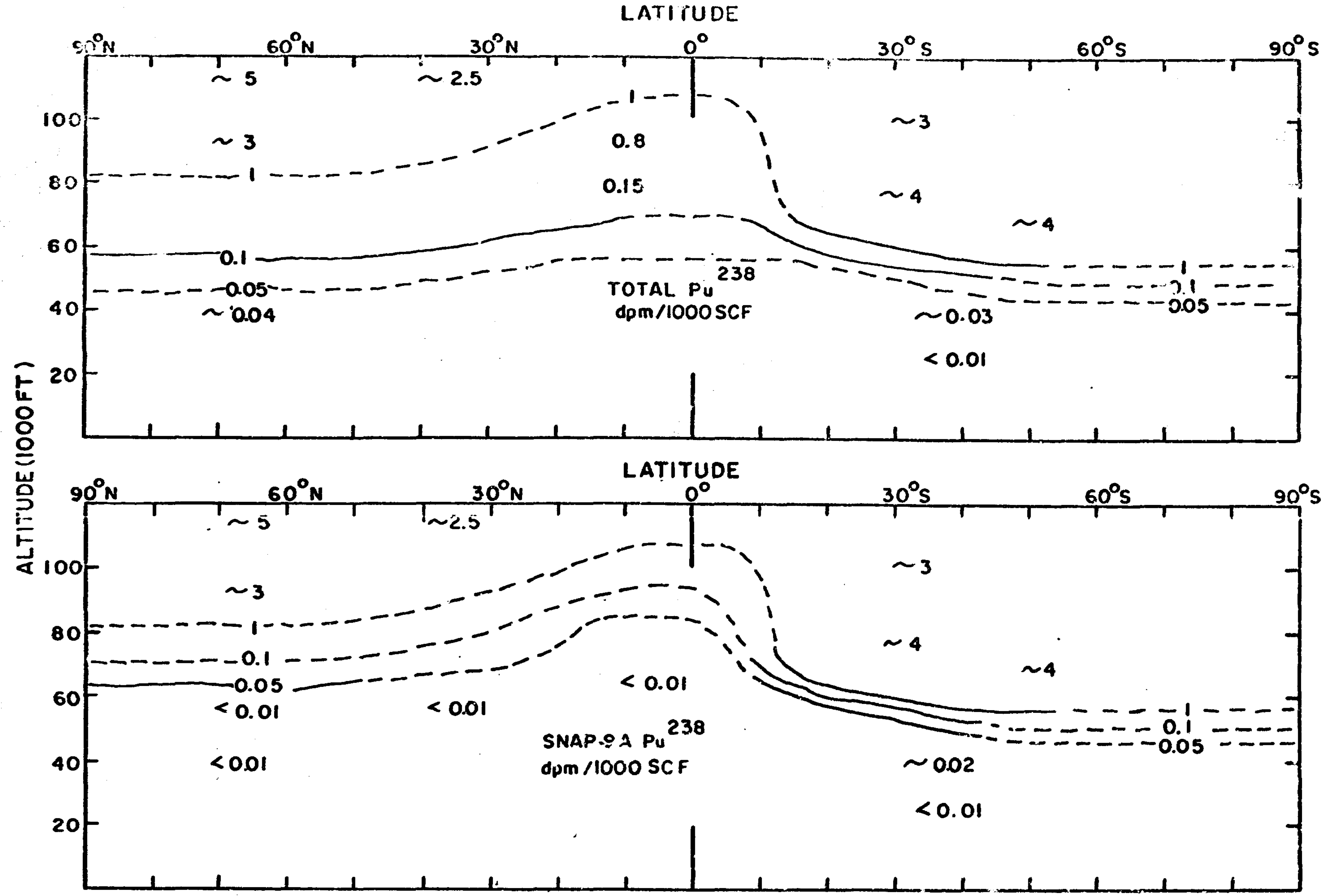


FIGURE 70. THE MEAN DISTRIBUTION OF TOTAL Pu^{238} AND OF SNAP-9A Pu^{239} DURING SEPT-DEC 1965

TABLE 5

THE STRATOSPHERIC BURDEN OF SNAP-9A PLUTONIUM-238 (IN KILOCURIES) DURING SEPTEMBER-DECEMBER, 1965

Altitude		Latitude				
10^3 ft	mb	90°-30°N	30°-0°N	0°-30°S	30°-90°S	Total
250-72	0-40	1.4	0.5	1.2	2.0	5.1
72-49	40-120	0.02	0.00	0.5	1.9	2.4
49-30	120-300	0.00	-	-	0.04	0.04
		1.4	0.5	1.7	3.9	7.5 kc.

TABLE 6

RESULTS OF PLUTONIUM ANALYSES OF SELECTED SAMPLES,
INCLUDING REANALYSES OF TWO ANOMALOUS SAMPLES

Sample Number	Collection Date	Latitude	Altitude (10^3 ft)	$\frac{Pu^{238}}{Pu^{239}}$	Total Pu^{239}	Total Pu^{238}	SNAP-9A Pu^{238}
SQ-7564	28 Apr 65	38°-55°S	62	0.04	1.08	0.04	<0.02
SX-7476	9 Nov 65	38°-55°S	63	3.34	1.51	5.04	4.99
SQ-7251	11 May 65	17°-36°S	53	0.13	0.54	0.07	0.05
SQ-7565	11 May 65	17°-36°S	53	0.04	0.58	0.02	<0.01
SQ-7264	23 Jun 65	17°-34°S	53	0.19	0.79	0.15	0.13
SQ-7566	23 Jun 65	17°-34°S	53	0.03	0.73	0.02	<0.02

TABLE 7

ESTIMATES OF SIZES OF HYPOTHETICAL SPHERICAL PARTICLES OF PuO_2

(1) $\frac{\Delta N}{\Delta t} = D = \lambda N,$

where $D = \frac{\Delta N}{\Delta t}$ = observed disintegration rate of Pu^{238}, in atoms/min.,

λ = decay constant of Pu^{238}, 1.48×10^{-8} $min.^{-1}$,

N = atoms of Pu^{238} in the sample.

(2) $N = A \frac{V \rho}{M}$,

where A = Avogadro's number = 6.02×10^{23} atoms/mole

V = volume of $Pu^{238}O_2$ particle, in cm^3

ρ = density of $Pu^{238}O_2$ = 11.46 g/cm^3

M = molecular weight of $Pu^{238}O_2$ = 270 g/mole

(3) $r^3 = \frac{V}{4.189}$

where r = radius of particles, in cm.

(4) $r^3 = \frac{V}{4.189} = \frac{NM}{4.189 \, A \rho} = \frac{DM}{4.189 \, \lambda A \rho} = \frac{270 \, D}{(4.189)(1.48 \times 10^{-8})(6.02 \times 10^{23})(11.46)}$

$r^3 = 0.631 \times 10^{-15} \, D$

For SQ-7264:

$D = (0.13 \text{ dpm}/1000 \text{ SCF}) (45.6 \times 10^3 \text{ SCF}) = 5.9 \text{ dpm}$

$r^3 = (0.631 \times 10^{-15}) (5.9) = 3.72 \times 10^{-15}$

$r = 1.5 \times 10^{-5} \text{ cm.} = 0.15 \, \mu$

For SQ-7251:

$D = (0.05 \text{ dpm}/1000 \text{ SCF}) (57.9 \times 10^3 \text{ SCF}) = 2.9 \text{ dpm}$

$r^3 = (0.631 \times 10^{-15}) (2.9) = 1.83 \times 10^{-15}$

$r = 1.2 \times 10^{-5} \text{cm.} = 0.12 \, \mu$

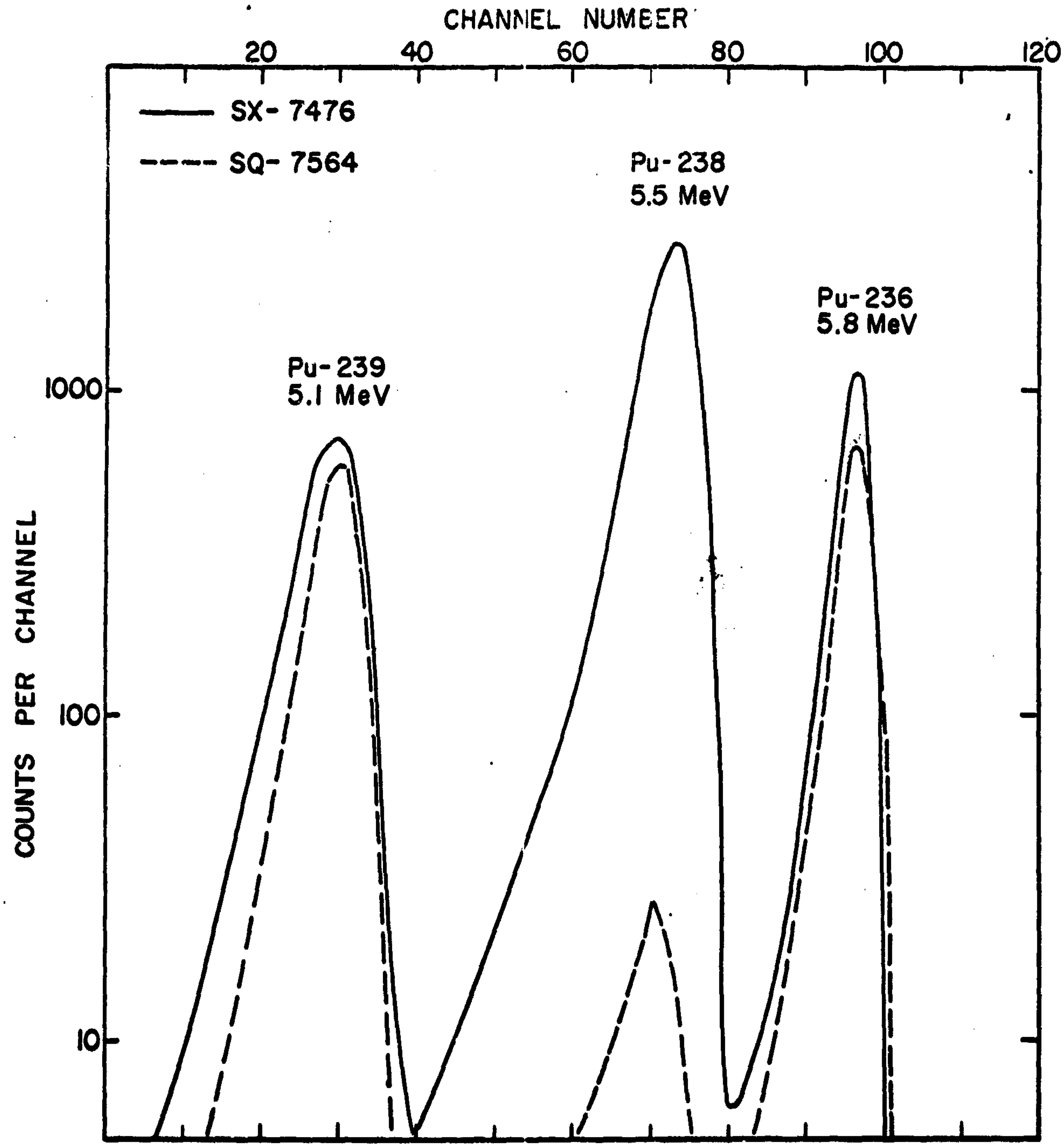


FIGURE 71. ALPHA SPECTRA OF SX-7476 CONTAINING SNAP-9A Pu-238 AND SQ-7564 CONTAINING ONLY WEAPONS Pu-238

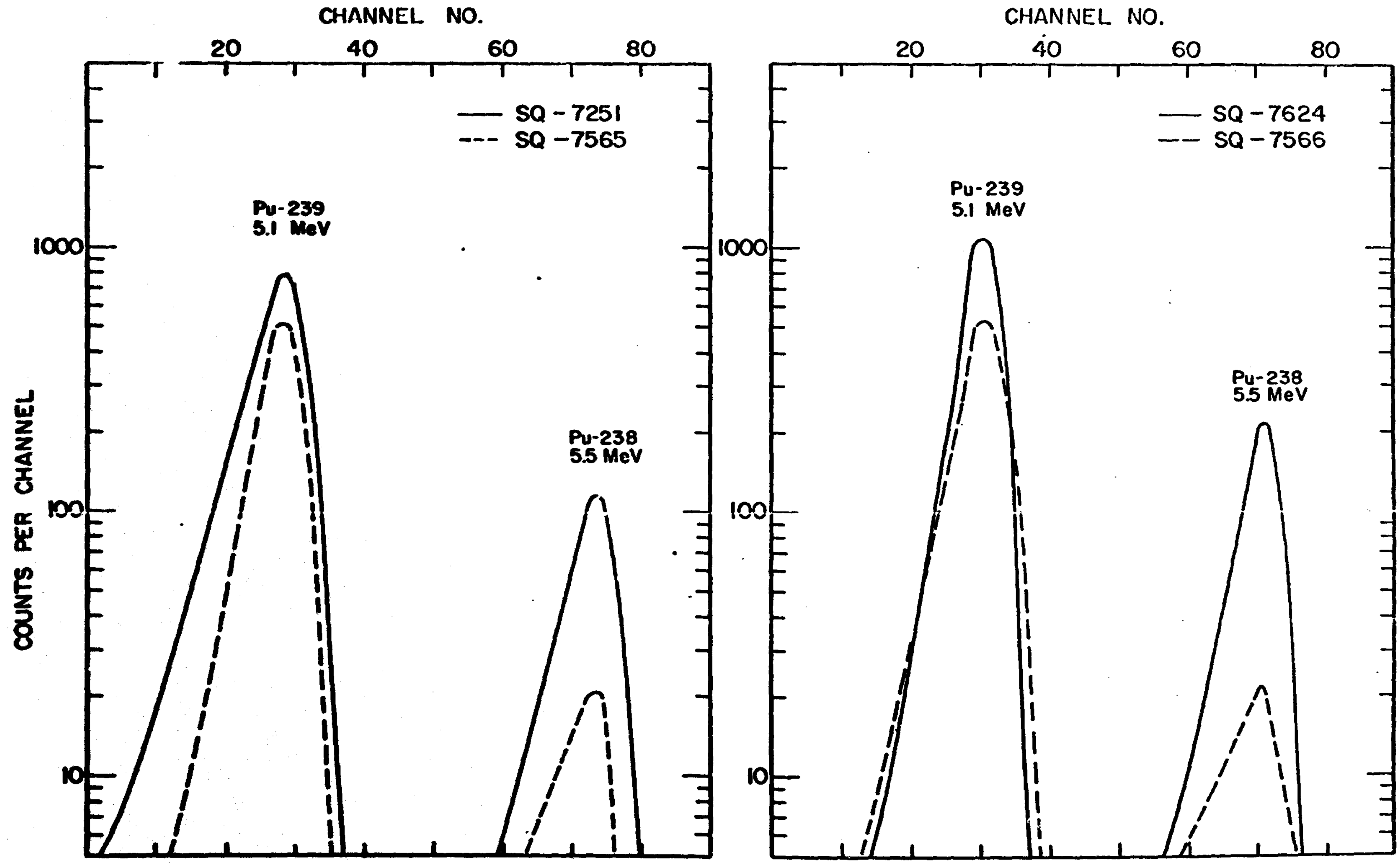


FIGURE 72. ALPHA SPECTRA OF DUPLICATE ANALYSES OF TWO ANOMALOUS SAMPLES

CHAPTER 6. THE STRATOSPHERIC DISTRIBUTION OF NATURAL RADIOACTIVITY

A number of radionuclides occur in the stratosphere as a result of natural processes. Several of these have been measured from time to time in Stardust filter samples. During 1963 to early 1965 a series of samples was measured for fcur products of cosmic ray induced spallation of atmospheric argon, nitrogen, and oxygen. A number of samples collected during 1957 to 1959 were measured for lead-210, and a set of samples collected in December, 1964 was measured for lead-210 and polonium-210. These nuclides are introduced into the atmosphere by the decay of radon-222 which escapes from soil, rocks, and bodies of water at the surface of the earth. The stratospheric concentrations of these nuclides have been measured in the expectation that their distributions would yield information on the directions and magnitudes of atmospheric motions.

The Stratospheric Distribution of Cosmic Ray Products

Beginning with samples collected in May, 1963 a number of Stardust samples were measured for the cosmic ray products beryllium-7, phosphorus-32, phosphorus-33 and sodium-22. It rapidly became evident that artificially produced sodium-22 was quite abundant in the stratosphere, as has been discussed in Chapter 4, making it impossible to determine the natural concentrations of that nuclide. The phosphorus samples were beta-counted with and without absorbers to determine the relative contributions of 1.71 Mev phosphorus-32 and 0.25 Mev phosphorus-33 beta particles. The measurements of phosphorus-32 were moderately successful, but those of phosphorus-33 were less so. An unsuccessful attempt was made to improve the phosphorus-33 data by eliminating counting through absorbers, and by using instead resolution of the phosphorus beta decay curve into two components (14.3 day phosphorus-32 and 25 day phosphorus-33) to distinguish between the contributions of the two radioisotopes. Unfortunately, by the time

the samples reached the laboratory and were processed only low levels of phosphorus activity remained, and this limited the accuracy of the measurements. The beryllium-7 and phosphorus-32 data are probably the most reliable of all of the activities of cosmic ray products measured. Nevertheless, some of the results for these nuclides appear to be too high or too low compared to the concentration range expected in the stratosphere.

Beryllium-7 and phosphorus-32 data for samples collected at three altitudes, 50, 60 and 65 thousand feet, at 65°-70°N are shown in Figure 73. The data suggest a possible seasonal trend, with high concentrations in the polar stratosphere during the summer months being replaced by low concentrations during the winter months. Beryllium-7 and phosphorus-32 data for equivalent altitudes in the tropical stratosphere and in the southern polar stratosphere do not show similar trends.

A correlation was sought between the appearance of low beryllium-7 and phosphorus-32 concentrations at 65°N during the winter and the shift in the stratospheric circulation which is characterized by the eastward movement onto the North American continent of a high pressure system which persists over the northern Pacific during the first half of the winter. The eastward migration of this high pressure system, associated with the disruption of the typical pole-centered low pressure system of the winter circulation, occurs at the time of the "explosive warmings" which usually affect the upper atmosphere over North America during late winter. During early 1962 and early 1963 the eastward movement of this high pressure system was accompanied by sudden decreases in the total beta activity of Stardust filter samples collected over Alaska. It was anticipated that such air motions would also produce sudden decreases in concentrations of cosmic ray products in the stratosphere in this region, since they would bring in northward moving air from low latitudes, where the production rates of cosmic ray products are relatively low. The beryllium-7 and phosphorus-32 concentrations and the

atmospheric temperatures at 65°N during October 1963 to March 1964 are plotted in Figure 74. The frequency of filter sample collection and the reliability of the radiochemical results were not sufficient to substantiate a correlation between changes in temperature and in nuclide concentrations.

The distributions of beryllium-7 and phosphorus-32 in the Stardust sampling corridor during December, 1963 are shown in Figure 75, and those during February, 1964 and February, 1965 are shown in Figures 76 and 77, respectively. The concentrations are plotted over horizontal lines representing the flight tracks of the sampling missions. The concentration isolines in the figure indicate the concentrations expected in a stagnant atmosphere, and are based on figures from Bhandari, et al[8]. The concentrations at 50 thousand feet and above in the northern polar stratosphere during February, 1964 were significantly lower than those in this region during December, 1963 indicating the possibility that an influx of air from lower latitudes had recently occurred. This was not true in February, 1965, but the data in Figure 73 suggest that a change in the pattern of the stratospheric circulation over Alaska may have occurred earlier, during January, 1965.

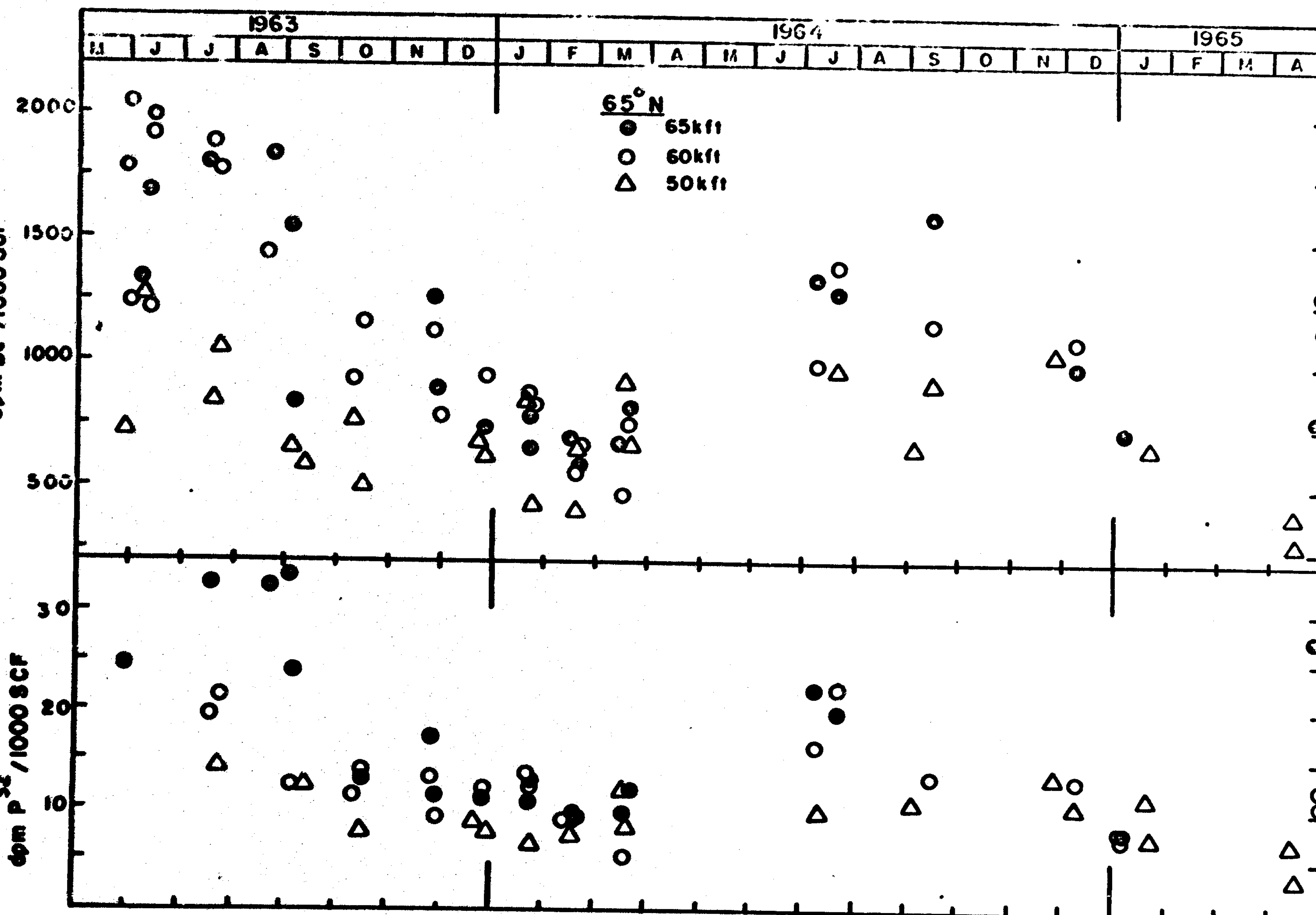


FIGURE 73. CONCENTRATIONS OF Be^7 AND P^{32} AT 65°-70°N

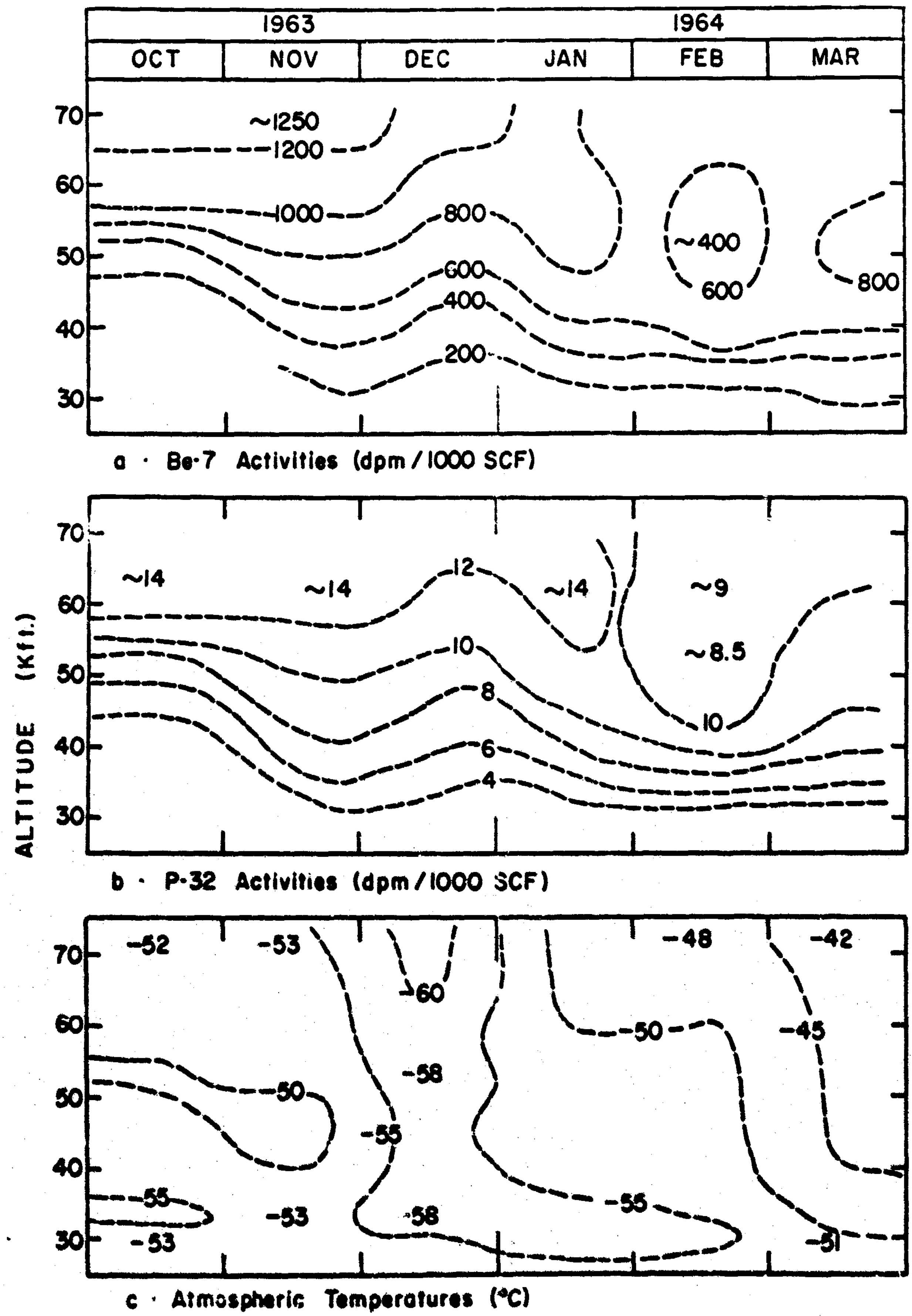


FIGURE 74. ACTIVITIES OF EERYLLIUM-7 AND PHOSPHORUS -32, AND TEMPERATURES AT 65°N DURING OCT. 1963 AND MAR. 1964

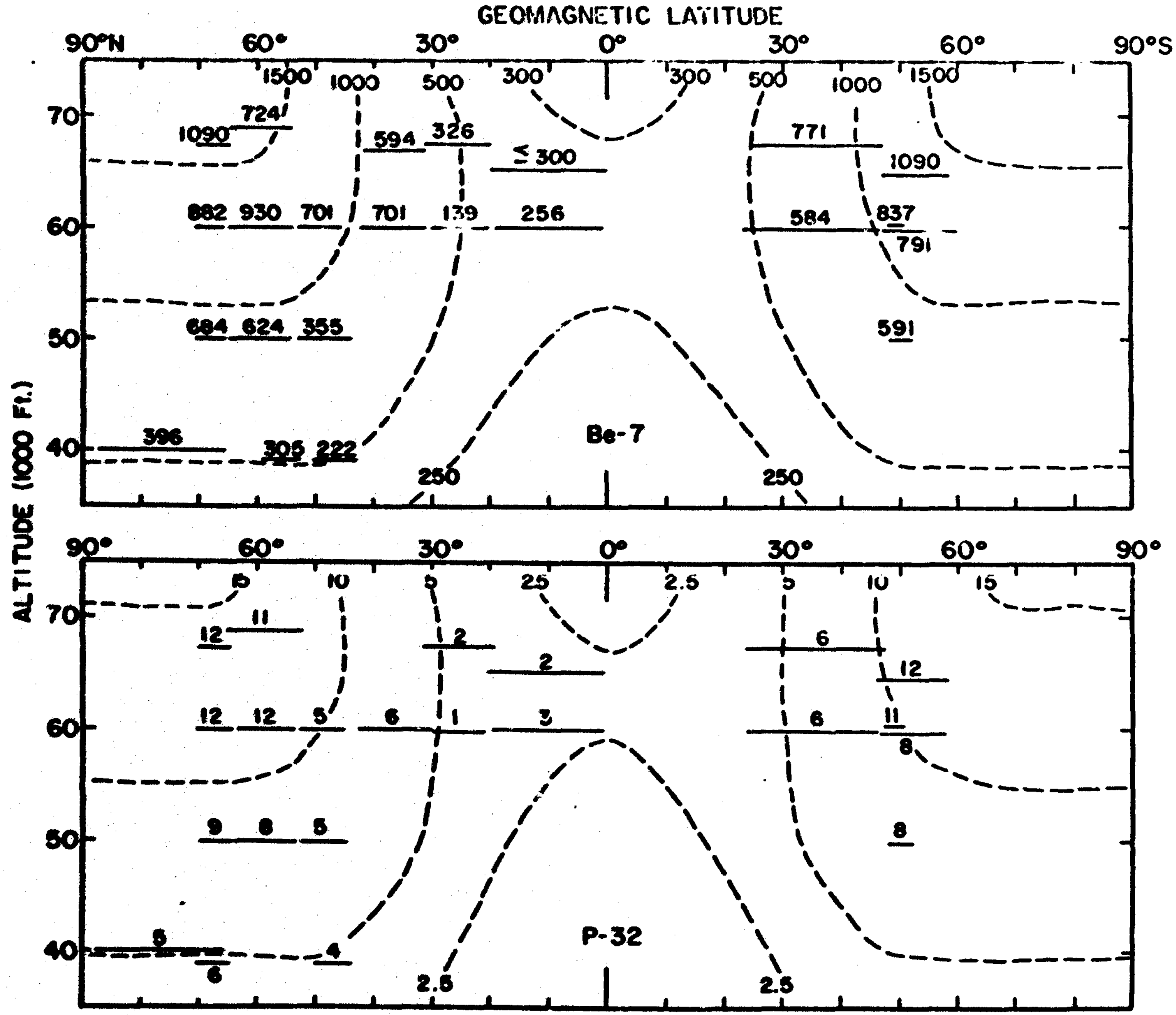


FIGURE 75. MEASUREMENTS OF Be-7 AND P-32 (dpm/1000 SCF) DURING DECEMBER 1963

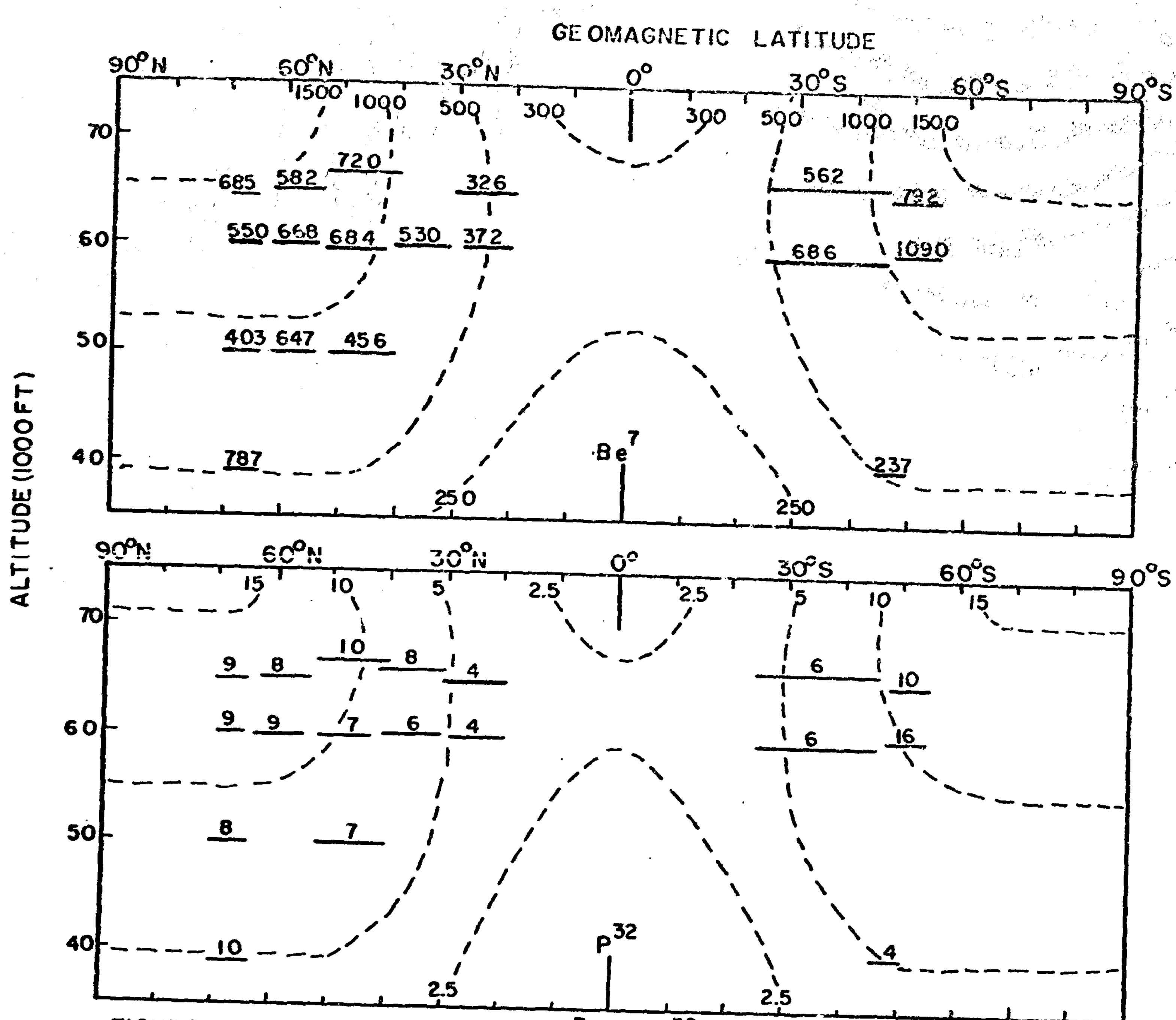


FIGURE 76. MEASUREMENTS OF Be^7 AND P^{32} (dpm/1000 SCF) DURING FEB 1964

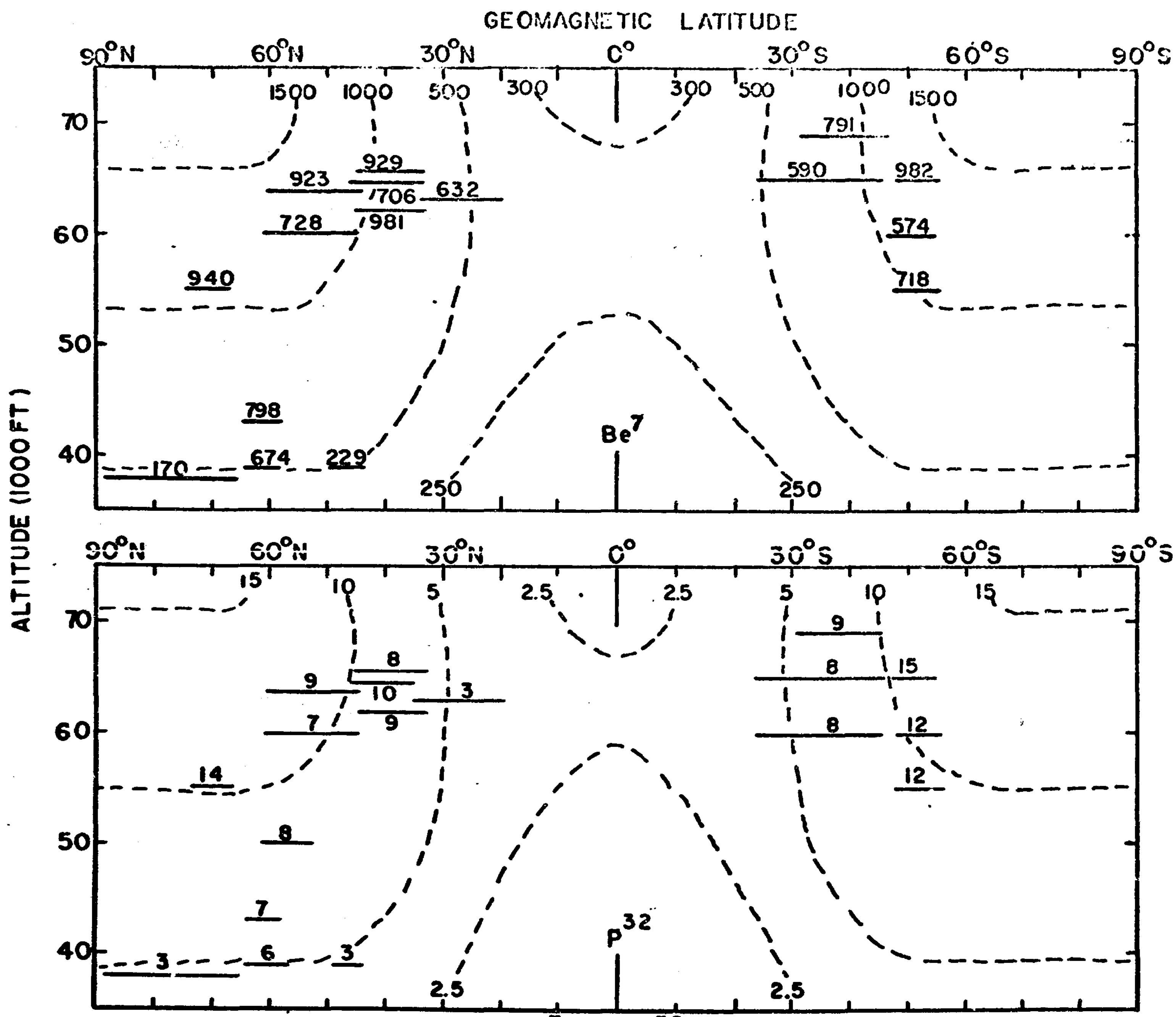


FIGURE 77 MEASUREMENTS OF Be^7 AND P^{32} (dpm/1000 SCF) DURING FEB 1965

The Stratospheric Distribution of Lead-210 and Polonium-210

Uranium is a fairly common trace constituent of rocks, soils and natural waters. The main isotope of uranium, uranium-238 (99.27% abundant), is radioactive, decaying through a series of intermediate radioactive daughter products ultimately to stable lead-206. The predominant decay series of uranium-238, which is summarized in Table 8, includes radon, which chemically behaves as a rare gas. When the uranium, or at least the immediate parent of radon, radium-226, is situated in rocks, soil or water close to the surface of the earth, some of the radon atoms produced by its decay may escape into the atmosphere during the few days which pass between their formation and their decay. When they do decay in the free atmosphere their daughter products will be ionized and will tend to undergo chemical reactions and to become attached to dust particles in the atmosphere. For the most part, the radon atoms which enter the atmosphere will remain in the lower troposphere, and their decay products will rapidly be washed out and returned to the surface of the earth. Some, however, will be carried into the upper troposphere or, perhaps, even into the lower stratosphere before they decay, and their daughter products may form within, or may be carried into, the stratosphere. As a result, low concentrations of lead-210 (radium-D), bismuth-210 (radium-E), and polonium-210 (radium-F) are found in filter samples of stratospheric air.

Results of measurements of lead-210 and polonium-210 in stratospheric air were explained by Burton and Stewart[16] in terms of the circulation of the atmosphere proposed by Brewer, and they concluded that approximately 200 days were required for air which entered the stratosphere at low latitudes to leave it at mid-latitudes. None of the subsequent studies which have included measurements of lead-210 in the stratosphere have confirmed the high concentrations reported by Burton and Stewart.

The stratospheric concentrations of lead-210 found by Rama and Honda[17] led them to the conclusion that the high concentrations found by Burton and Stewart "appear to be due to some local causes." Based on their own data Rama and Honda concluded that the residence time of lead-210 in the stratosphere "is long enough to permit the processes of mixing to make the concentration more or less independent of altitude and latitude."

In the Ninth Quarterly Report on Project Stardust, DASA-1309, we concluded, on the basis of measurements of samples collected during June, 1961 to April, 1962, that lead-210 concentrations decreased with altitude above the tropopause layer, and that the Po^{210}/Pb^{210} activity ratio increased with height in the troposphere and reached an equilibrium value of about 1.0 in the stratosphere. It was believed that the available data were compatible with the model of Jacobs and Andre[18], which calculated the vertical distribution of radon-222 and its daughter products in the atmosphere by means of a solution of the diffusion equation, with an assumed vertical profile of the coefficient of turbulent diffusion.

Bhandari et al[8], on the basis of published values of stratospheric lead-210 concentrations, concluded that the lead-210 content of stratospheric air between 55° and 75° latitude is lower than the lead-210 content of other atmospheric regions, and that the highest lead-210 concentrations occur in the equatorial stratosphere. They attribute the low values in the polar stratosphere to removal of aerosols from that region by gravitational settling, with a mean time of settling of 30 months. They suggest intrusions of tropospheric air into the tropical stratosphere, perhaps by highly turbulent vertical mixing associated with thunderstorms, as the cause of the high lead-210 concentrations in the tropical stratosphere.

To obtain more data on the distribution of lead-210 in the stratosphere, measurements were made on a series of filter samples collected during 1957 to 1959 as part of Project HASP. These measurements were made during 1961 to 1963, originally by separating lead from the samples, allowing bismuth-210 to grow in,

and then beta counting the bismuth-210. Some results, however, suggested that some samples might be affected by contamination with beta emitters from newer samples which were collected during and after the 1961 and 1962 series of weapons tests, and were being analyzed in the same laboratory. As a result, measurements of polonium-210 in the HASP samples were begun also to verify the results of the bismuth-210 analyses. Since the samples were 3 to 6 years old the polonium-210 was assumed to be in radioactive equilibrium with the lead-210. The results of the bismuth-210 and polonium-210 measurements are summarized in Table 9, and a frequency distribution of measured concentrations is shown in Figure 78. The agreement between bismuth-210 and polonium-210 results is not good. The polonium-210 data are considered the more reliable since they show less scatter, and since the polonium-210 analyses were less subject to the effects of contamination. Where it seemed possible and important to eliminate erroneous data, polonium-210 or bismuth-210 values which appeared too high or too low were rejected, perhaps in some cases somewhat arbitrarily. The lead-210 values obtained from Table 9 are included in Table 10 with flight data and results of strontium-90 and cerium-144 analyses of the samples. Besides the measured strontium-90 values, Table 11 also contains estimated concentrations of strontium-90 in the samples, based on data from analyses performed during Project HASP.

The mean distribution of lead-210 in the stratosphere during October, 1957 to July, 1959, based on the data in Table 10, is summarized in Table 11 and is plotted in Figure 79. The data points used to draw the isolines in Figure 79 are plotted in the figure, with different symbols used to indicate the number of samples represented. The details of the distribution are uncertain due to the scarcity of samples in some regions, and to the apparent low level of accuracy of some of the measurements. Nevertheless there is general agreement between this distribution and that reported in DASA-1309 for Stardust samples collected during

June - September, 1961. It is also consistent with measurements of lead-210 in the stratosphere reported by the U.S. Weather Bureau[19].

The chief feature of the distribution shown in Table 11 and Figure 79 is the occurrence of a layer of maximum concentration in the lower stratosphere, at a height of about 45 thousand feet in the polar stratosphere, but sloping upward toward the equator to a height of 60 thousand feet in the tropical stratosphere. Above this layer concentrations decrease with height. The highest concentrations found were in the equatorial stratosphere, but some of the high activity samples contained debris from a September, 1958 United Kingdom weapon test, so artificial lead-210 may be represented.

Lead-210 and polonium-210 measurements were made on a group of samples collected in December, 1964 to check earlier estimates of the stratospheric distribution of the Po^{210}/Pb^{210} activity ratio. To minimize the likelihood that samples would contain interfering beta activity, the samples chosen were those collected in the tropical and southern polar stratosphere. The results of the analyses are summarized in Table 12. Lead-210 results given for samples SR-6900, SR-6901 and SR-7734 are based on recent analyses, but polonium-210 results for the first two are based on analyses performed in early 1965.

The flight tracks of the sampling missions which collected these lead-polonium samples and the approximate location of the tropopause are plotted in Figure 80, together with the lead-210 and polonium-210 data.

The available results indicate that the Po^{210}/Pb^{210} activity ratio in the upper troposphere is well below 1.0, but that in the lower stratosphere this ratio equals or even exceeds 1.0. The two samples from 60 and 65 thousand feet at 38°-47°S seem to have Po^{210}/Pb^{210} ratios below 1.0, though admittedly the data are of questionable reliability. Some additional analyses of lead-210 and polonium-210 are being performed on samples collected during late 1965 and early 1966 to

obtain better evidence on the distribution of these nuclides within the stratosphere. The final results of these analyses will be discussed in a later report.

TABLE 8

THE PREDOMINANT DECAY SERIES OF URANIUM-238

Nuclide	Name	Half Life	Mode of Decay
U^{238}	Uranium I	4.51×10^9 years	α
Th^{234}	Uranium X_1	24.10 days	β^-
Pa^{234}	Uranium X_2	1.18 minutes	β^- (99.4%)
U^{234}	Uranium II	2.48×10^5 years	α
Th^{230}	Ionium	7.6×10^4 years	α
Ra^{226}	Radium	1.62×10^3 years	α
Rn^{222}	Radon	3.823 days	α
Po^{218}	Radium A	3.05 minutes	α (99.97%)
Pb^{214}	Radium B	26.8 minutes	β^-
Bi^{214}	Radium C	19.7 minutes	β^- (99.96%)
Po^{214}	Radium C	1.64×10^{-4} seconds	α
Pb^{210}	Radium D	22 years	β^-
Bi^{210}	Radium E	5.0 days	β^- (~100%)
Po^{210}	Radium F	138.4 days	α
Pb^{206}	Radium G	stable	-

TABLE 9

RESULTS OF BISMUTH-210 AND POLONIUM-210 ANALYSIS OF HASP SAMPLES
(DATA WHICH ARE BELIEVED TO BE INCORRECT AND HAVE BEEN REJECTED ARE PLACED IN PARENTHESES)

Sample Number	Collection Date	$\frac{\text{dpm Bi}^{210}}{1000 \text{ SCF}}$	$\frac{\text{dpm Po}^{210}}{1000 \text{ SCF}}$	Po^{210}/Bi^{210}	$\frac{\text{dpm Pb}^{210}}{1000 \text{ SCF}}$
ST-83	29 Aug 57	0.67A	0.51A	0.76	0.59
ST-84	4 Oct 57	0.89A	($\leq$0.04)	($\leq$0.04)	0.89
ST-1	16 Oct 57	0.59B	-	-	0.59
ST-85	8 Nov 57	(1.32B)	0.32C	(0.24)	0.32
ST-86	12 Nov 57	0.72A	0.33A	0.46	0.52
ST-87	12 Nov 57	0.67B	0.26B	0.39	0.46
ST-88	20 Nov 57	0.51B	0.44B	0.87	0.48
ST-4	22 Nov 57	(1.45A)	-	-	-
ST-5	22 Nov 57	0.50C	-	-	0.50
ST-89	22 Nov 57	1.02A	0.63B	0.62	0.82
ST-90	26 Nov 57	-	0.55A	-	0.55
ST-6	26 Nov 57	0.15C	-	-	0.15
ST-91	26 Nov 57	0.68A	0.50A	0.73	0.59
ST-92	3 Dec 57	0.50B	0.21B	0.42	0.36
ST-93	3 Dec 57	0.72B	0.51B	0.71	0.62
ST-94	3 Dec 57	0.72B	0.34B	0.47	0.53
ST-95	14 Dec 57	0.32B	0.35B	1.09	0.34
ST-96	14 Dec 57	(0.09D)	0.49A	(5.44)	0.49
ST-97	14 Dec 57	0.37B	0.44A	1.18	0.40
ST-98	17 Dec 57	0.28B	0.46A	1.62	0.37
ST-99	10 Jan 58	0.55B	0.49A	0.89	0.52
ST-8	10 Jan 58	0.43A	-	-	0.43
ST-100	10 Jan 58	0.22B	0.15B	0.65	0.18
ST-10	24 Jan 58	0.32B	-	-	0.32
ST-11	24 Jan 58	0.26B	-	-	0.26
ST-12	31 Jan 58	0.42A	-	-	0.42
ST-13	31 Jan 58	0.35C	-	-	0.35
ST-102	31 Jan 58	0.50B	0.31A	0.62	0.40
ST-103	4 Feb 58	0.41B	0.42A	1.01	0.42
ST 104	7 Feb 58	0.77B	0.35A	0.45	0.56
ST-14	7 Feb 58	0.32C	-	-	0.32
ST-15	7 Feb 58	0.22C	-	-	0.22
ST-105	21 Feb 58	0.39A	0.37B	0.95	0.38
ST-17	21 Feb 58	0.34A	-	-	0.34
ST-106	26 Feb 58	0.4.	0.38B	0.93	0.40
ST-107	26 Feb 58	0.88B	0.26B	0.29	0.57
ST-108	1 Mar 58	0.54A	0.46A	0.86	0.50
ST-109	1 Mar 58	0.22C	0.28B	1.23	0.25
ST-18	1 Mar 58	0.26A	-	-	0.26
ST-110	5 Mar 58	-	0.59A	-	0.59
ST-111	5 Mar 58	0.65C	0.40D	0.61	0.52
ST-112	11 Mar 58	0.57A	0.31A	0.54	0.44
ST-19	28 Mar 58	0.30A	-	-	0.30
ST-20	28 Mar 58	0.31A	-	-	0.31

TABLE 9 (continued)

RESULTS OF BISMUTH-210 AND POLONIUM-210 ANALYSIS OF HASP SAMPLES
(DATA WHICH ARE BELIEVED TO BE INCORRECT AND HAVE BEEN REJECTED ARE PLACED IN PARENTHESES)

Sample Number	Collection Date	dpm Bi^{210} / 1000 SCF	dpm Po^{210} / 1000 SCF	Po^{210}/Bi^{210}	dpm Pb^{210} / 1000 SCF
ST-21	28 Mar 58	0.31B	-	-	0.31
ST-22	28 Mar 58	0.35A	-	-	0.35
ST-23	1 Apr 58	0.15A	-	-	0.15
ST-24	1 Apr 58	0.26A	-	-	0.26
ST-25	1 Apr 58	0.75A	-	-	0.75
ST-26	4 Apr 58	0.36A	-	-	0.36
ST-27	4 Apr 58	0.35A	-	-	0.35
ST-28	8 Apr 58	0.22A	-	-	0.22
ST-29	8 Apr 58	0.42A	-	-	0.42
ST-113	8 Apr 58	0.49A	0.37A	0.76	0.43
ST-30	8 Apr 58	0.31A	-	-	0.31
ST-31	15 Apr 58	0.37B	-	-	0.37
ST-32	15 Apr 58	0.33A	-	-	0.33
ST-33	15 Apr 58	0.59B	-	-	0.59
ST-34	25 Apr 58	0.24A	-	-	0.24
ST-115	2 May 58	0.30B	0.35A	1.15	0.32
ST-116	2 May 58	0.36A	0.27A	0.75	0.31
ST-35	6 May 58	0.39B	-	-	0.39
ST-36	6 May 58	0.53A	-	-	0.53
ST-37	6 May 58	0.42A	-	-	0.42
ST-38	6 May 58	0.28B	-	-	0.28
ST-40	6 Jun 58	0.17B	-	-	0.17
ST-41	6 Jun 58	0.38A	-	-	0.38
ST-118	20 Jun 58	0.39B	0.26B	0.66	0.32
ST-43	24 Jun 58	0.64A	-	-	0.64
ST-44	24 Jun 58	0.58B	-	-	0.58
ST-45	1 Jul 58	0.21C	-	-	0.21
ST-46	1 Jul 58	0.26C	-	-	0.26
ST-120	4 Jul 58	0.72B	0.47A	0.66	0.60
ST-121	4 Jul 58	0.44B	0.41A	0.94	0.42
ST-47	8 Jul 58	0.62B	-	-	0.62
ST-48	12 Sep 58	0.31A	0.25B	0.79	0.28
ST-49	12 Sep 58	0.74C	0.58A	0.78	0.66
ST-122	19 Sep 58	(2.26A)	0.58C	(0.26)	0.58
ST-50	19 Sep 58	0.60B	-	-	0.60
ST-51	23 Sep 58	0.61B	0.47B	0.77	0.54
ST-52	23 Sep 58	0.58A	-	-	0.58
ST-53	23 Sep 58	-	0.40A	-	0.40
ST-54	27 Sep 58	0.42A	0.50A	1.20	0.46
ST-55	27 Sep 58	0.28A	(0.69A)	(2.46)	0.28
ST-56	30 Sep 58	0.46A	-	-	0.46
ST-57	30 Sep 58	0.42A	-	-	0.42
ST-58	1 Oct 58	0.26B	-	-	0.26
ST-124	2 Oct 58	-	(0.06C)	-	-

TABLE 9 (continued)

RESULTS OF BISMUTH-210 AND POLONIUM-210 ANALYSIS OF HASP SAMPLES
(DATA WHICH ARE BELIEVED TO BE INCORRECT AND HAVE BEEN REJECTED ARE PLACED IN PARENTHESES)

Sample Number	Collection Date	dpm Bi^{210} / 1000 SCF	dpm Po^{210} / 1000 SCF	Po^{210}/Bi^{210}	dpm Pb^{210} / 1000 SCF
ST-125	3 Oct 58	-	0.75A	-	0.75
ST-59	7 Oct 58	0.68B	-	-	0.68
ST-126	7 Oct 58	1.63C	1.93C	0.84	1.78
ST-128	10 Oct 58	0.34C	0.30B	0.88	0.32
ST-60	10 Oct 58	0.42B	-	-	0.42
ST-129	14 Oct 58	(1.60B)	0.71A	(0.44)	0.71
ST-61	14 Oct 58	0.19B	0.21A	1.06	0.20
ST-62	17 Oct 58	0.78A	-	-	0.78
ST-64	19 Oct 58	0.24B	-	-	0.24
ST-131	21 Oct 58	-	0.50A	-	0.50
ST-132	21 Oct 58	(1.27B)	0.40C	(0.32)	0.40
ST-133	22 Oct 58	-	0.31B	-	0.31
ST-135	29 Oct 58	1.05B	0.89A	0.84	0.97
ST-136	29 Oct 58	0.56B	0.57A	1.02	0.56
ST-71	7 Nov 58	0.51A	-	-	0.51
ST-72	7 Nov 58	0.31B	-	-	0.31
ST-137	15 Nov 58	0.69B	0.70A	1.02	0.70
ST-74	15 Nov 58	0.29C	-	-	0.29
ST-75	16 Nov 58	0.23B	-	-	0.23
ST-138	18 Nov 58	1.01B	0.24A	0.24	0.63
ST-140	18 Nov 58	-	0.16C	-	0.16
ST-141	20 Nov 58	-	0.44B	-	0.44
ST-76	21 Nov 58	0.42B	-	-	0.42
ST-143	21 Nov 58	0.34B	0.34B	1.02	0.34
ST-77	22 Nov 58	0.24B	0.56A	2.31	0.40
ST-78	25 Nov 58	0.81B	-	-	0.81
ST-79	25 Nov 58	0.66A	-	-	0.66
ST-80	25 Nov 58	0.26B	0.32A	1.24	0.29
ST-81	28 Nov 58	0.77B	-	-	0.77
ST-82	28 Nov 58	0.16B	0.46A	2.83	0.31
ST-145	3 Dec 58	0.33B	0.32A	0.95	0.32
ST-146	3 Dec 58	0.39B	0.31B	0.79	0.35
ST 147	3 Dec 58	0.46B	0.46B	0.98	0.46
ST-148	3 Dec 58	0.93D	0.46B	0.50	0.70
ST-149	3 Dec 58	0.45B	0.54B	1.21	0.50
ST-150	3 Dec 58	0.90B	0.60B	0.67	0.75
ST-151	9 Dec 58	0.64B	0.47A	0.74	0.55
ST-152	12 Dec 58	0.61A	0.47A	0.76	0.54
ST-153	12 Dec 58	0.46B	0.67A	1.45	0.56
ST-154	12 Dec 58	0.17C	0.53A	3.04	0.35
ST-155	12 Dec 58	(1.22C)	0.18D	(0.14)	0.18
ST 156	16 Dec 58	0.54B	0.43B	0.80	0.48
ST-157	16 Dec 58	0.55B	0.41A	0.74	0.48
ST-158	16 Dec 58	0.35B	0.37A	1.05	0.36

TABLE 9 (continued)

RESULTS OF BISMUTH-210 AND POLONIUM-210 ANALYSIS OF HASP SAMPLES
(DATA WHICH ARE BELIEVED TO BE INCORRECT AND HAVE BEEN REJECTED ARE PLACED IN PARENTHESES)

Sample Number	Collection Date	dpm Bi^{210} / 1000 SCF	dpm Po^{210} / 1000 SCF	Po^{210}/Bi^{210}	dpm Pb^{210} / 1000 SCF
ST-159	16 Dec 58	0.23B	0.30B	1.30	0.26
ST-160	19 Dec 58	0.32C	0.34B	1.04	0.33
ST-161	19 Dec 58	0.32C	0.33B	1.04	0.32
ST-162	19 Dec 58	0.57B	0.43A	0.76	0.50
ST-163	19 Dec 58	0.36B	0.18B	0.51	0.27
ST-164	23 Dec 58	0.28B	0.28A	1.02	0.28
ST-165	23 Dec 58	0.35B	0.34A	0.98	0.34
ST-166	23 Dec 58	0.66B	0.45B	0.68	0.56
ST-167	23 Dec 58	0.26C	0.30A	1.13	0.28
ST-173	6 Jan 59	-	0.42A	-	0.42
ST-174	9 Jan 59	-	0.49A	-	0.49
ST-175	9 Jan 59	-	0.37A	-	0.37
ST-176	13 Jan 59	-	0.41A	-	0.41
ST-177	13 Jan 59	-	0.36A	-	0.36
ST-178	16 Jan 59	-	0.36B	-	0.36
ST-179	19 Jan 59	-	0.29B	-	0.29
ST-180	22 Jan 59	-	0.37A	-	0.37
ST-181	25 Jan 59	-	0.32A	-	0.32
ST-182	28 Jan 59	-	0.30A	-	0.30
ST-183	3 Feb 59	-	0.36B	-	0.36
ST-184	6 Feb 59	-	0.40A	-	0.40
ST-185	6 Feb 59	-	0.40A	-	0.40
ST-186	6 Feb 59	-	0.22B	-	0.22
ST-187	10 Feb 59	-	0.36A	-	0.36
ST-188	14 Feb 59	-	0.44A	-	0.44
ST-189	14 Feb 59	-	0.36A	-	0.36
ST-190	14 Feb 59	-	0.16C	-	0.16
ST-191	20 Feb 59	-	0.34A	-	0.34
ST-192	20 Feb 59	-	0.37B	-	0.37
ST-193	20 Feb 59	-	0.39A	-	0.39
ST-194	24 Feb 59	-	0.45A	-	0.45
ST-195	24 Feb 59	-	0.31B	-	0.31
ST-196	24 Feb 59	-	0.38A	-	0.38
ST-197	6 Mar 59	-	0.38A	-	0.38
ST-198	6 Mar 59	-	0.34B	-	0.34
ST-199	10 Mar 59	-	0.28A	-	0.28
ST-200	10 Mar 59	-	0.40A	-	0.40
ST-201	13 Mar 59	-	0.19B	-	0.19
ST-202	17 Mar 59	-	0.36A	-	0.36
ST-203	17 Mar 59	-	0.45A	-	0.45
ST-204	17 Mar 59	-	0.37A	-	0.37
ST-205	24 Mar 59	-	0.34A	-	0.34
ST-206	27 Mar 59	-	0.19B	-	0.19
ST-207	1 Apr 59	-	0.42A	-	0.42
ST-208	3 Apr 59	-	0.43A	-	0.43
ST-209	3 Apr 59	-	0.42A	-	0.42

TABLE 9 (continued)

RESULTS OF BISMUTH-210 AND POLONIUM-210 ANALYSIS OF HASP SAMPLES
(DATA WHICH ARE BELIEVED TO BE INCORRECT AND HAVE BEEN REJECTED ARE PLACED IN PARENTHESES)

Sample Number	Collection Date	dpm Bi^{210} / 1000 SCF	dpm Po^{210} / 1000 SCF	Po^{210}/Bi^{210}	dpm Pb^{210} / 1000 SCF
ST-210	7 Apr 59	-	0.33A	-	0.33
ST-211	10 Apr 59	-	0.34A	-	0.34
ST-212	10 Apr 59	-	0.32B	-	0.32
ST-213	10 Apr 59	-	0.07D	-	0.07
ST-214	14 Apr 59	-	0.20A	-	0.20
ST-215	17 Apr 59	-	0.40B	-	0.40
ST-216	17 Apr 59	-	0.39A	-	0.39
ST-217	17 Apr 59	-	0.30A	-	0.30
ST-218	19 Apr 59	-	0.26A	-	0.26
ST-219	21 Apr 59	-	0.30A	-	0.30
ST-220	21 Apr 59	-	0.16B	-	0.16
ST-221	24 Apr 59	-	0.22C	-	0.22
ST-222	5 May 59	-	0.32B	-	0.32
ST-224	8 May 59	-	0.45C	-	0.45
ST-225	12 May 59	-	0.42B	-	0.42
ST-226	12 May 59	-	0.17C	-	0.17
ST-227	15 May 59	-	0.39A	-	0.39
ST-228	15 May 59	-	0.34A	-	0.34
ST-229	20 May 59	-	0.28A	-	0.28
ST-230	20 May 59	-	0.32A	-	0.32
ST-231	20 May 59	-	0.40A	-	0.40
ST-232	24 May 59	-	0.25A	-	0.25
ST-233	24 May 59	-	0.23B	-	0.23
ST-234	26 May 59	-	0.37A	-	0.37
ST-235	26 May 59	-	0.35A	-	0.35
ST-236	29 May 59	-	0.45A	-	0.45
ST-237	2 Jun 59	-	0.20	-	0.20
ST-238	9 Jun 59	-	0.45A	-	0.45
ST-239	12 Jun 59	-	0.38A	-	0.38
ST-240	16 Jun 59	-	0.36A	-	0.36
ST-241	16 Jun 59	-	0.25B	-	0.25
ST-242	16 Jun 59	-	0.46B	-	0.46
ST-244	26 Jun 59	-	0.31B	-	0.31
ST-245	26 Jun 59	-	0.55A	-	0.55
ST-246	1 Jul 59	-	0.24A	-	0.24
ST-247	7 Jul 59	-	0.14B	-	0.14
ST-248	10 Jul 59	-	(1.20A)	-	-
ST-249	14 Jul 59	-	0.40A	-	0.40
ST-250	21 Jul 59	-	(0.06A)	-	-
ST-251	21 Jul 59	-	0.45A	-	0.45
ST-252	24 Jul 59	-	0.33A	-	0.33
ST-253	24 Jul 59	-	0.23A	-	0.23
ST-254	28 Jul 59	-	0.57A	-	0.57
ST-255	28 Jul 59	-	0.54A	-	0.54
ST-256	28 Jul 59	-	0.34A	-	0.34
ST-257	28 Jul 59	-	0.61B	-	0.61

TABLE 10

RESULTS OF ANALYSIS OF HASP SAMPLES

Sample Number	Collection Date	Latitude	Altitude (10^3 ft)	Altitude (km)	Activities (dpm/1000 SCF) Total β	Sr^{90} (est.)	Sr^{90} (meas)	Ce^{144}	Pb^{210}
ST-83	29 Aug 57	32°N	56	17	17,700	58	78A	(2,450A)	0.59
ST-84	4 Oct 57	32°N	64	20	43,000	179	188A	2,200A	0.89
ST-1	16 Oct 57	28°-14°N	56	17	30,000	72	-	-	0.59
ST-85	8 Nov 57	65°-44°N	56	17	66,000	158	178A	2,630A	0.32
ST-86	12 Nov 57	34°-22°N	66	20	190,000	253	222A	4,960A	0.52
ST-87	12 Nov 57	22°N-2°S	66	20	270,000	282	314A	6,350A	0.46
ST-88	20 Nov 57	45°-22°N	68	21	20,000	132	128A	1,400A	0.48
ST-4	22 Nov 57	65°-45°N	47	14	92,000	130	-	-	-
ST-5	22 Nov 57	44°-22°N	62	19	210,000	250	-	-	0.50
ST-89	22 Nov 57	16°N-7°S	63	19	5,300,000	1,150	1,270A	37,800A	0.82
ST-90	26 Nov 57	64°-51°N	44	13	174,000	206	(25A)	5,440A	0.55
ST-6	26 Nov 57	44°-16°N	59	18	132,000	134	-	-	0.15
ST-91	26 Nov 57	16°N-7°S	59	18	92,000	77	84A	2,340A	0.59
ST-92	3 Dec 57	67°-44°N	59	18	145,000	196	206A	4,920A	0.36
ST-93	3 Dec 57	39°-22°N	55	17	20,000	32	32A	782A	0.62
ST-94	3 Dec 57	21°-11°N	57	17	12,800	19	(5.3A)	518A	0.53
ST-95	14 Dec 57	66°-44°N	59	18	129,000	200	216A	5,410A	0.34
ST-96	14 Dec 57	44°-33°N	56	17	123,000	175	158A	5,080A	0.49
ST-97	14 Dec 57	33°-22°N	56	17	34,000	59	62A	1,480A	0.40
ST-98	17 Dec 57	44°-22°N	53	16	76,000	110	113A	2,940A	0.37
ST-99	10 Jan 58	55°-44°N	56	17	92,000	207	230A	4,750A	0.52
ST-8	10 Jan 58	44°-22°N	52	16	52,000	110	-	-	0.43
ST-100	10 Jan 58	21°N-6°S	52	16	4,400	5.0	6.0A	-	0.18
ST-10	24 Jan 58	65°-45°N	50	15	108,000	218	-	-	0.32
ST-11	24 Jan 58	15°N-5°S	48	15	810	1.6	-	-	0.26
ST-12	31 Jan 58	65°-46°N	47	14	61,000	154	-	-	0.42
ST-13	31 Jan 58	42°-23°N	44	13	13,700	≤ 38	-	-	0.35
ST-102	31 Jan 58	20°N-4°S	44	13	690	1.0	(112A)	-	0.40

TABLE 10 (continued)

RESULTS OF ANALYSIS OF HASP SAMPLES

Sample Number	Collection Date	Latitude	Altitude (10^3 ft)	Altitude (km)	Activities (dpm/1000 SCF) Total β	Sr^{90} (est.)	Sr^{90} (meas)	Ce^{144}	Pb^{210}
ST-103	4 Feb 58	37°-16°N	65	20	70,000	264	(446A)	5,840A	0.42
ST-104	7 Feb 58	67°-44°N	55	17	32,000	210	131A	3,780A	0.56
ST-14	7 Feb 58	44°-26°N	55	17	13,200	81	-	-	0.32
ST-15	7 Feb 58	16°N-6°S	55	17	≤810	≤3.5	-	-	0.22
ST-105	21 Feb 58	45°-33°N	55	17	29,000	192	-	3,680A	0.38
ST-17	21 Feb 58	38°-16°N	68	21	15,800	124	-	-	0.34
ST-106	26 Feb 58	45°-33°N	45	14	20,000	122	135A	2,920A	0.40
ST-107	26 Feb 58	38°-16°N	65	20	17,000	110	130A	2,400A	0.57
ST-108	1 Mar 58	55°-33°N	55	17	9,800	70	85A	1,510A	0.50
ST-109	1 Mar 58	33°-22°N	55	17	290	2.0	3.0A	-	0.25
ST-18	1 Mar 58	16°N-7°S	55	17	710	4.3	-	-	0.26
ST-110	5 Mar 58	67°-44°N	55	17	21,000	175	192A	2,960A	0.59
ST-111	5 Mar 58	16°N-6°S	62	19	53,000	282	406D	7,140A	0.52
ST-112	11 Mar 58	38°-16°N	67	20	-	171	203A	2,950A	0.44
ST-19	28 Mar 58	61°-44°N	55	17	492,000	339	-	(87A)	0.30
ST-20	28 Mar 58	44°-21°N	55	17	(3,100)	70	-	-	0.31
ST-21	28 Mar 58	38°-16°N	67	20	28,000	157	-	-	0.31
ST-22	28 Mar 53	16°N-7°S	55	17	≤470	2.5	-	-	0.35
ST-23	1 Apr 58	68°-44°N	64	20	44,000	114	108A	1,960A	0.15
ST-24	1 Apr 58	44°-16°N	65	20	139,000	192	214A	4,340A	0.26
ST-25	1 Apr 58	16°N-6°S	65	20	20,000	128	132A	2,550A	0.75
ST-26	4 Apr 58	44°-22°N	45	14	68,000	133	-	-	0.36
ST-27	4 Apr 58	44°-16°N	57	17	230,000	210	249A	5,180A	0.35
ST-28	8 Apr 58	66°-44°N	55	17	200,000	219	-	-	0.22
ST-29	8 Apr 58	44°-22°N	55	17	34,000	60	78A	1,840A	0.42
ST-113	8 Apr 58	38°-16°N	65	20	21,000	137	196A	2,710A	0.43
ST-30	8 Apr 58	16°N-7°S	55	17	370	20	-	-	0.31
ST-31	15 Apr 58	67°-44°N	55	17	110,000	206	253A	5,690A	0.37
ST-32	15 Apr 58	44°-21°N	54	16	152,000	190	176A	3,740A	0.33
ST-33	15 Apr 58	16°N-6°S	55	17	630	2.0	-	-	0.59
ST-34	25 Apr 58	38°-16°N	67	20	23,000	108	144A	2,080A	0.24

TABLE 1C (continued)

RESULTS OF ANALYSIS OF HASP SAMPLES

Sample Number	Collection Date	Latitude	Altitude (10^3 ft)	Altitude (km)	Activities (dpm/1000 SCF) Total β	Sr^{90}(est.)	Sr^{90}(meas)	Ce^{144}	Pb^{210}
ST-115	2 May 58	67°-44°N	55	17	86,000	199	259A	5,640A	0.32
ST-116	2 May 58	38°-16°N	66	20	18,800	128	166A	2,390A	0.31
ST 35	6 May 58	67°-44°N	55	17	124,000	235	-	5,800A	0.39
ST-36	6 May 58	44°-22°N	55	17	34,000	63	-	1,920A	0.53
ST-37	6 May 58	38°-16°N	67	20	34,000	146	183A	2,980A	0.42
ST-38	6 May 58	16°N-5°S	56	17	5,700	8.3	-	-	0.28
ST-40	6 Jun 58	19°N	46	14	1,170	2.5	-	-	0.17
ST-41	6 Jun 58	19°N	56	17	99,000	61	50A	1,090A	0.38
ST-42	6 Jun 58	19°N	65	20	73,000	128	157A	3,200A	-
ST-118	20 Jun 58	19°-18°N	67	20	73,000	158	231A	3,360A	0.32
ST-43	24 Jun 58	38°-16°N	60	18	75,000	173	171A	3,200A	0.64
ST-44	24 Jun 58	16°N-6°S	60	18	115,000	84	-	1,600A	0.58
ST-45	1 Jul 58	19°N	56	17	19,700	41	-	-	0.21
ST-46	1 Jul 58	19°N	67	20	100,000	200	-	-	0.26
ST-120	4 Jul 58	39°-16°N	60	18	55,000	119	163A	2,990A	0.60
ST-121	4 Jul 58	16°-7°N	60	18	88,000	78	101A	1,890A	0.42
ST-47	8 Jul 58	19°N	56	17	360,000	194	189A	5,530A	0.62
ST-48	12 Sep 58	75°-71°N	66	20	2,700	74	73A	739A	0.28
ST-49	12 Sep 58	10°N-18°S	66	20	210,000	419	381A	5,920A	0.66
ST-122	19 Sep 58	3°S-16°S	60	18	96,000	289	170A	2,930A	0.58
ST-50	19 Sep 58	16°S-35°S	60	18	7,500	83	-	-	0.60
ST-51	23 Sep 58	38°-21°N	60	18	36,900	132	157A	2,160A	0.54
ST-52	23 Sep 58	21°N-6°S	60	18	250,000	248	250A	4,700A	0.58
ST-53	23 Sep 58	35°-40°N	40	12	4,600	11	10A	171A	0.40
ST-54	27 Sep 58	71°N	50	15	13,200	153	138A	2,270A	0.46
ST-55	27 Sep 58	71°N	64	20	2,800	60	86A	812A	0.28
ST-56	30 Sep 58	16°S-35°S	60	18	12,400	73	77A	1,000A	0.46
ST-57	30 Sep 58	35°S-57°S	60	18	13,100	84	86A	1,170A	0.42

TABLE 10 (continued)

RESULTS OF ANALYSIS OF HASP SAMPLES

Sample Number	Collection Date	Latitude	Altitude (10^3 ft)	Altitude (km)	Activities (dpm/1000 SCF) Total β	Sr^{90} (est.)	Sr^{90} (meas)	Ce^{144}	Pb^{210}
ST-58	1 Oct 58	71°N	60	18	7,500	159	134A	1,660A	0.26
ST-124	2 Oct 58	73°-71°N	50	15	1,400	(12)	31A	397A	-
ST-125	3 Oct 58	21°N-8°S	60	18	390,000	364	408A	8,560A	0.75
ST-59	7 Oct 58	38°-16°N	60	18	45,200	146	184A	2,580A	0.68
ST-126	7 Oct 58	35°-40°S	50	15	15,000	(49)	89A	2,070A	1.78
ST-127	7 Oct 58	45°-45°S	65	20	26,000	200	250A	7,020A	-
ST-128	10 Oct 58	38°-16°N	60	18	29,000	151	119A	2,150A	0.32
ST-60	10 Oct 58	35°-57°S	60	18	10,500	74	85A	1,230A	0.42
ST-129	14 Oct 58	16°N-6°S	60	18	740,000	897	510A	19,440A	0.71
ST-61	14 Oct 58	35°-45°S	33	10	5,200	22	-	-	0.20
ST-130	17 Oct 58	38°-23°N	60	18	150,000	315	368A	8,210A	-
ST-62	17 Oct 58	22°N-8°S	60	18	510,000	636	479A	12,380A	0.78
ST-64	19 Oct 58	44°-20°N	50	15	14,000	48	-	-	0.24
ST-131	21 Oct 58	35°-43°S	60	18	9,600	75	90A	(3,140A)	0.50
ST-132	21 Oct 58	35°-44°S	65	20	13,000	111	134A	(51A)	0.40
ST-133	22 Oct 58	44°N	40	12	950	(34)	2.8A	308A	0.31
ST-134	22 Oct 58	44°N	70	21	103,000	187	141A	4,720A	-
ST-67	23 Oct 58	56°-44°N	55	17	96,000	202	162A	3,050A	-
ST-135	29 Oct 58	44°-27°N	50	15	21,000	56	64A	1,960A	0.97
ST-69	29 Oct 58	44°-20°N	57	17	670,000	532	370A	12,250A	-
ST-136	29 Oct 58	38°-20°N	55	17	42,000	105	131A	3,520A	0.56
ST-71	7 Nov 58	17°N-22°S	60	18	151,000	307	296A	6,910A	0.51
ST-72	7 Nov 58	29°-57°S	60	18	9,400	68	80A	1,080A	0.31
ST-137	15 Nov 58	38°-17°N	60	18	400,000	482	489A	14,650A	0.70
ST-74	15 Nov 58	29°-57°S	60	18	7,500	68	-	-	0.29
ST-75	16 Nov 58	42°-20°N	50	15	22,000	42	-	-	0.23
ST-138	18 Nov 58	66°-44°N	50	15	240,000	270	267A	5,860A	0.63
ST-139	18 Nov 58	55°-44°N	60	18	87,000	202	217A	4,600A	-
ST-140	18 Nov 58	66°-44°N	65	20	8,900	76	68A	1,040A	0.16
ST 141	20 Nov 58	55°-45°N	60	18	52,000	184	211A	-	0.44
ST-142	20 Nov 58	55°-45°N	65	20	23,000	143	156A	2,650A	-
ST-76	21 Nov 58	10°-29°S	60	18	37,000	102	-	-	0.42
ST-143	21 Nov 58	29°-57°S	60	18	12,000	55	87A	3,010A	0.34
ST-77	22 Nov 58	44°-20°N	58	18	103,000	207	213A	4,440A	0.40

TABLE 10 (continued)

RESULTS OF ANALYSIS OF HASP SAMPLES

Sample Number	Collection Date	Latitude	Altitude (10^3 ft)	Altitude (km)	Activities (dpm/1000 SCF) Total β	Sr^{90} (est.)	Sr^{90} (meas)	Ce^{144}	Pb^{210}
ST-78	25 Nov 58	38°-22°N	60	18	92,000	245	218A	5,010A	0.81
ST-79	25 Nov 58	22°N-6°S	60	18	88,000	229	205A	5,020A	0.66
ST-80	25 Nov 58	35°-40°S	34	10	1,700	3.0	-	-	0.29
ST-144	25 Nov 58	35°-40°S	55	17	40,000	167	195A	2,770A	-
ST-81	28 Nov 58	16°N-8°S	60	18	55,000	144	129A	3,530A	0.77
ST-82	28 Nov 58	17°-57°S	60	18	12,000	66	92A	1,230A	0.31
ST-145	3 Dec 58	35°-40°S	30	9	760	≤5.0	(68A)	(1,910A)	0.32
ST-146	3 Dec 58	35°-40°S	35	11	1,000	6.0	(36A)	(428A)	0.35
ST-147	3 Dec 58	40°-41°S	40	12	4,300	26	33A	(4,040A)	0.46
ST-148	3 Dec 58	35°-40°S	45	14	8,500	28	39A	1,900A	0.70
ST-149	3 Dec 58	42°S	50	15	12,000	60	(4.8A)	2,940A	0.50
ST-150	3 Dec 58	35°-40°S	55	17	17,000	64	(21A)	1,630A	0.75
ST-151	9 Dec 58	38°-28°N	50	15	20,000	43	43A	1,580A	0.55
ST-152	12 Dec 58	38°-17°N	60	18	76,000	238	202A	5,240A	0.54
ST 153	12 Dec 58	16°N-8°S	60	18	73,000	188	197A	5,110A	0.56
ST-154	12 Dec 58	10°-35°S	60	18	11,000	55	61A	1,310A	0.35
ST-155	12 Dec 58	35°-57°S	60	18	6,200	62	77A	3,990A	0.18
ST 156	16 Dec 58	38°-27°N	55	17	56,000	143	-	3,210A	0.48
ST-157	16 Dec 58	25° 5°N	55	17	5,000	11	13B	440A	0.48
ST-158	16 Dec 58	2°-35°S	55	17	5,400	(16)	32B	3,240A	0.36
ST-159	16 Dec 58	35°-57°S	55	17	9,600	61	72A	629A	0.26
ST-160	19 Dec 58	38°-16°N	65	20	93,000	358	385A	7,270A	0.33
ST-161	19 Dec 58	16°N-8°S	65	20	130,000	490	504A	11,300A	0.32
ST-162	19 Dec 58	10°-35°S	65	20	9,600	58	79A	1,220A	0.50
ST-163	19 Dec 58	35°-57°S	65	20	5,500	48	72A	1,060A	0.27
ST 164	23 Dec 58	34°-40°S	30	9	390	3.0	2.0A	56B	0.28
ST 165	23 Dec 58	35°-40°S	35	11	1,100	2.0	2.2A	121A	0.34
ST-166	23 Dec 58	35°-40°S	45	14	3,400	16	22A	525A	0.56
ST-167	23 Dec 58	42° 43°S	50	15	6,500	20	25A	578A	0.28

TABLE 10 (continued)

RESULTS OF ANALYSIS OF HASP SAMPLES

Sample Number	Collection Date	Latitude	Altitude (10^3 ft)	Altitude (km)	Activities (dpm/1000 SCF) Total β	Sr^{90} (est.)	Sr^{90} (meas)	Ce^{144}	Pb^{210}
ST-173	6 Jan 59	29°-57°S	50	15	7,900	41	48A	966A	0.42
ST-174	9 Jan 59	16°-8°S	60	18	43,000	155	160A	4,130A	0.49
ST-175	9 Jan 59	10°-57°S	60	18	8,000	67	63A	1,070A	0.37
ST-176	13 Jan 59	40°S	40	12	2,100	16	19A	345A	0.41
ST 177	13 Jan 59	41°-43°S	50	15	6,300	36	50A	799A	0.36
ST-178	16 Jan 59	10°-51°S	65	20	9,200	84	102A	1,360A	0.36
ST-179	19 Jan 59	38°-20°N	65	20	50,000	276	291A	5,030A	0.29
ST-180	22 Jan 59	35°-57°S	50	15	5,700	31	46A	(23A)	0.37
ST-181	25 Jan 59	44°-20°N	60	18	52,000	230	254A	4,420A	0.32
ST-182	28 Jan 59	35°-51°S	55	17	5,900	44	57A	883A	0.30
ST-183	3 Feb 59	41°-57°S	50	15	5,300	41	48A	761A	0.36
ST-184	6 Feb 59	38°-10°N	60	18	37,000	151	150A	3,740A	0.40
ST-185	6 Feb 59	10°-29°S	60	18	5,900	39	51A	874A	0.40
ST-186	6 Feb 59	40°-57°S	60	18	5,000	59	66A	942A	0.22
ST-187	10 Feb 59	41°-57°S	55	17	6,000	42	62A	830A	0.36
ST-188	14 Feb 59	32°N°-8°S	65	20	47,000	217	270A	4,800A	0.44
ST-189	14 Feb 59	10°-35°S	65	20	8,400	72	80A	1,130A	0.36
ST-190	14 Feb 59	35°-57°S	65	20	4,700	57	63A	847A	0.16
ST-191	20 Feb 59	27°N-8°S	65	20	51,000	213	254A	4,490A	0.34
ST-192	20 Feb 59	16°-35°S	65	20	7,800	58	75A	1,200A	0.37
ST-193	20 Feb 59	35°-57°S	48	15	5,000	25	34A	592A	0.39
ST-194	24 Feb 59	38°N-8°S	66	20	57,000	269	305A	5,230A	0.45
ST-195	24 Feb 59	16°-35°S	66	20	6,600	54	67A	1,020A	0.31
ST-196	24 Feb 59	35°-57°S	55	17	6,800	(24)	55A	801A	0.38
ST-197	6 Mar 59	32°N-2°S	65	20	41,000	192	225A	3,840A	0.38
ST-198	6 Mar 59	16°-35°S	67	20	6,400	58	69A	1,040A	0.34
ST-199	10 Mar 59	38°-16°N	60	18	29,000	139	171A	3,750A	0.28
ST-200	10 Mar 59	35°-57°S	55	17	4,800	47	50A	1,160A	0.40
ST 201	13 Mar 59	35°-51°S	65	20	6,090	68	70A	1,890A	0.19
ST-202	17 Mar 59	32°N-8°S	65	20	32,000	167	166A	3,860A	0.36
ST-203	17 Mar 59	2°-28°S	65	20	14,000	89	100A	3,010A	0.45
ST-204	17 Mar 59	35°-55°S	40	12	1,200	8.0	8.7A	347A	0.37
ST-205	24 Mar 59	35°-57°S	55	17	4,200	38	37A	1,440A	0.34
ST-206	27 Mar 59	16°-52°S	65	20	6,000	66	61A	1,300A	0.19

TABLE 10 (continued)

RESULTS OF ANALYSIS OF HASP SAMPLES

Sample Number	Collection Date	Latitude	Altitude (10^3 ft)	Altitude (km)	Activities (dpm/1000 SCF) Total β	Sr^{90} (est.)	Sr^{90} (meas)	Ce^{144}	Pb^{210}
ST-207	1 Apr 59	32°N-8°S	68	21	45,000	246	278A	5,210A	0.42
ST-208	3 Apr 59	35°-40°S	40	12	1,000	8.0	8.9A	276B	0.43
ST-209	3 Apr 59	35°-40°S	50	15	3,500	25	16A	649A	0.42
ST-210	7 Apr 59	29°-52°S	55	17	4,400	50	49A	929A	0.33
ST-211	10 Apr 59	38°-10°N	65	20	31,000	196	246A	3,790A	0.34
ST-212	10 Apr 59	35°-40°S	60	18	5,100	51	64A	1,160A	0.32
ST-213	10 Apr 59	35°-40°S	66	20	4,700	58	67A	1,970A	0.07
ST-214	14 Apr 59	90°-60°N	36	11	47,000	171	190A	5,000A	0.20
ST 215	17 Apr 59	38°-27°N	55	17	19,000	114	(1,280A)	2,340A	0.40
ST-216	17 Apr 59	35°-40°S	40	12	810	6.0	-	-	0.39
ST-217	17 Apr 59	35°-40°S	50	15	3,100	(16)	31A	399A	0.30
ST-218	19 Apr 59	44°-20°N	60	18	25,000	155	187A	4,360A	0.26
ST-219	21 Apr 59	66°-44°N	50	15	20,000	100	141A	2,550A	0.30
ST-220	21 Apr 59	66°-44°N	60	18	26,000	250	205A	3,660A	0.16
ST-221	24 Apr 59	35°-40°S	65	20	4,400	63	66A	(110B)	0.22
ST-222	5 May 59	16°-35°S	65	20	7,400	74	88A	1,370A	0.32
ST-224	8 May 59	35°-40°S	66	20	5,000	62	(147B)	(3,140A)	0.45
ST-225	12 May 59	29°-35°S	55	17	3,200	36	40A	621A	0.42
ST-226	12 May 59	35°-57°S	60	20	3,800	59	64A	1,280A	0.17
ST 227	15 May 59	35°-40°S	40	12	2,000	19	20A	435A	0.39
ST-228	15 May 59	35°-40°S	50	15	3,600	40	43A	662A	0.34
ST-229	20 May 59	68°-44°N	55	17	17,000	136	148A	1,940A	0.28
ST-230	20 May 59	10°-35°S	65	20	10,000	101	101A	1,850A	0.32
ST-231	20 May 59	35°-57°S	45	14	1,800	20	21A	382A	0.40
ST-232	24 May 59	44°-20°N	60	18	16,000	127	134A	2,660A	0.25
ST-233	24 May 59	44°-20°N	66	20	20,000	232	221A	-	0.23
ST-234	26 May 59	12°-32°S	60	18	3,800	44	51A	-	0.37
ST-235	26 May 59	46°-57°S	51	16	3,100	51	49A	1,420A	0.35
ST-236	29 May 59	35°-40°S	40	12	1,600	22	18A	-	0.45

TABLE 10 (continued)

RESULTS OF ANALYSIS OF HASP SAMPLES

Sample Number	Collection Date	Latitude	Altitude (10^3 ft)	Altitude (km)	Activities (dpm/1000 SCF) Total β	Sr^{90} (est.)	Sr^{90} (meas)	Ce^{144}	Pb^{210}
ST-237	2 Jun 59	31°-35°S	65	20	4,500	59	(17A)	1,430A	0.20
ST-238	9 Jun 59	23°-55°S	45	14	1,000	11	10A	-	0.45
ST-239	12 Jun 59	38°-4°N	64	20	19,000	192	(120A)	-	0.38
ST-240	16 Jun 59	60°-47°N	55	17	16,000	163	149A	-	0.36
ST-241	16 Jun 59	68°-47°N	65	20	21,000	264	246A	-	0.25
ST-242	16 Jun 59	35°-50°S	55	17	2,700	36	39A	863A	0.46
ST-244	26 Jun 59	35°-40°S	40	12	1,100	15	(87A)	(1,320A)	0.31
ST-245	26 Jun 59	35°-40°S	50	15	1,700	21	20A	562A	0.55
ST-246	1 Jul 59	38°N-2°S	65	20	19,000	208	(7.2A)	(212A)	0.24
ST-247	7 Jul 59	2°-28°S	65	20	10,000	129	(234A)	(1,080A)	0.14
ST-248	10 Jul 59	35°-40°S	50	15	1,700	25	(581A)	(7,220A)	-
ST-249	14 Jul 59	27°-4°N	68	21	24,000	286	268A	2,580A	0.40
ST-250	21 Jul 59	67°-44°N	45	14	7,600	70	(5.0A)	(84A)	-
ST-251	21 Jul 59	15°-35°S	60	18	3,700	45	44A	998A	0.45
ST-252	24 Jul 59	68°-45°N	55	17	16,000	162	184A	2,250A	0.33
ST-253	24 Jul 59	62°-45°N	65	20	16,000	221	215A	2,730A	0.23
ST-254	28 Jul 59	35°S	43	13	≤440	4.0	5.2A	(270A)	0.57
ST-255	28 Jul 59	35°S	48	15	870	12	12A	390A	0.54
ST-256	28 Jul 59	35°S	53	16	2,600	50	46A	643A	0.34
ST-257	28 Jul 59	35°S	60	18	5,200	74	83A	1,260A	0.61

Note: Letters following nuclide concentrations represent counting statistics:

A = 0-5% C = 10-20%
B = 5-10% D = 20-50%

Data which are believed to be incorrect and have been rejected are placed in parentheses.

TABLE 11

THE MEAN DISTRIBUTION OF LEAD 210 (DPM/1000 SCF), DURING OCTOBER 1957 - JULY 1959
(THE NUMBER OF SAMPLES WHICH EACH VALUE REPRESENTS FOLLOWS IT IN PARENTHESES)

Latitude	Altitude (1000 feet) 30	35	40	45	50	55	60	65	70
90°N		0.20 (1)							
85°N		0.20 (1)							
80°N		0.20 (1)							
75°N		0.20 (1)						0.28 (1)	
70°N		0.20 (1)			0.46 (1)		0.26 (1)	0.28 (1)	
65°N		0.20 (1)		0.42 (1)	0.42 (3)	0.38 (9)	0.29 (3)	0.19 (3)	
60°N		0.20 (1)		0.48 (2)	0.42 (3)	0.37 (11)	0.29 (3)	0.20 (4)	
55°N				0.48 (2)	0.42 (3)	0.39 (13)	0.32 (4)	0.20 (4)	
50°N				0.42 (1)	0.42 (3)	0.39 (13)	0.32 (4)	0.20 (4)	
45°N			0.31 (1)	0.40 (1)	0.42 (3)	0.39 (13)	0.32 (4)	0.18 (3)	0.48 (1)
40°N			0.40 (1)	0.37 (3)	0.47 (4)	0.40 (10)	0.31 (6)	0.27 (3)	0.48 (1)
35°N			0.40 (1)	0.37 (3)	0.48 (5)	0.43 (14)	0.46 (16)	0.35 (16)	0.41 (2)
30°N				0.36 (2)	0.48 (5)	0.42 (14)	0.46 (16)	0.39 (21)	0.41 (3)
25°N				0.36 (2)	0.30 (3)	0.43 (12)	0.46 (16)	0.37 (21)	0.41 (4)
20°N				0.28 (2)	0.22 (3)	0.44 (9)	0.49 (17)	0.36 (23)	0.39 (3)
15°N				0.40 (1)	0.22 (2)	0.40 (9)	0.60 (13)	0.44 (12)	0.41 (2)
10°N				0.40 (1)	0.22 (2)	0.36 (7)	0.60 (13)	0.46 (13)	0.41 (2)
5°N				0.40 (1)	0.22 (2)	0.36 (7)	0.64 (11)	0.43 (11)	0.41 (2)
0°N				0.40 (1)	0.22 (2)	0.34 (6)	0.64 (11)	0.47 (11)	0.42 (1)
5°S					0.22 (2)	0.34 (7)	0.63 (11)	0.48 (9)	0.42 (1)
10°S						0.36 (1)	0.42 (5)	0.40 (7)	
15°S						0.36 (1)	0.42 (7)	0.40 (7)	
20°S						0.36 (1)	0.42 (10)	0.33 (11)	
25°S				0.45 (1)		0.36 (1)	0.41 (9)	0.33 (11)	
30°S				0.45 (1)	0.42 (1)	0.37 (3)	0.38 (10)	0.34 (9)	
35°S	0.30 (2)	0.30 (4)	0.39 (6)	0.54 (5)	0.57 (9)	0.40 (10)	0.40 (13)	0.28 (16)	
40°S	0.30 (2)	0.30 (4)	0.40 (8)	0.53 (4)	0.52 (11)	0.40 (8)	0.33 (11)	0.25 (10)	
45°S		0.20 (1)	0.37 (1)	0.42 (2)	0.36 (5)	0.35 (8)	0.32 (9)	0.22 (6)	
50°S			0.37 (1)	0.42 (2)	0.38 (5)	0.35 (8)	0.32 (9)	0.22 (6)	
55°S			0.37 (1)	0.42 (2)	0.38 (5)	0.35 (5)	0.32 (9)	0.20 (3)	

TABLE 12

RESULTS OF ANALYSES OF DECEMBER, 1964 SAMPLES FOR LEAD-210 AND POLONIUM-210

Sample Number	Collection Date	Latitude	Altitude (10^3 ft)	Altitude (km)	Activities (dpm/1000 SCF) Total β	Sr^{90}	Pb^{210}	Po^{210}	Po^{210}/Pb^{210}
SR-6893	1 Dec 64	40°-45°S	15	4.6	39	1.1A	Lost	$\leq$0.03	-
SR-6894	1 Dec 64	40°-45°S	25	7.6	135	4.3A	0.17A	0.06A	0.34
SR-6895	8 Dec 64	37°-38°S	30	9.2	225	7.3A	0.09B	Lost	-
SR-6896	8 Dec 64	37°-38°S	35	10.7	83	2.9A	0.22A	Lost	-
SR-6897	8 Dec 64	37°S	40	12.2	1,110	39A	0.24C	0.24B	1.00
SR-6898	8 Dec 64	37°-38°S	45	13.7	1,900	42A	0.20D	0.34B	1.71
SR-6899	8 Dec 64	38°-47°S	55	16.8	4,900	150A	0.31C	0.41B	1.32
SR-6900	8 Dec 64	38°-47°S	60	18.3	4,180	144A	0.28C	0.13D	0.46
SR-6901	8 Dec 64	38°-47°S	65	19.8	5,030	119A	0.40C	0.20D	0.50
SR-7734	8 Dec 64	38°-47°S	66	20.1	4,170	106A	0.12B	-	-
SR-6902	21 Dec 64	9°N-10°S	55	16.8	322	7.5A	0.19B	0.15A	0.77
SR-6903	21 Dec 64	9°N-10°S	65	19.8	12,100	273A	0.44B	0.46A	1.03
SR-6904	22 Dec 64	37°-38°S	30	9.2	91	2.9A	0.23A	$\leq$0.01	$\leq$0.04
SR-6905	22 Dec 64	37°-38°S	35	10.7	123	2.9A	0.23A	0.07A	0.30
SR-6906	22 Dec 64	15°-37°S	55	16.8	1,310	37A	0.33A	0.23A	0.68
SR-6907	22 Dec 64	15°-87°S	65	19.8	6,100	111A	Lost	$\leq$0.28	-
SR-6908	23 Dec 64	9°N-10°S	60	18.3	5,900	152A	0.54A	0.53A	0.97
SR-6909	23 Dec 64	37°S	40	12.2	1,900	59A	0.18B	0.24A	1.33
SR-6910	23 Dec 64	37°S	45	13.7	2,190	55A	0.25A	0.24A	0.97

Note: Letters following nuclide concentrations represent counting statistics:

A = 0-5%　　C = 10-20%
B = 5-10%　　D = 20-50%

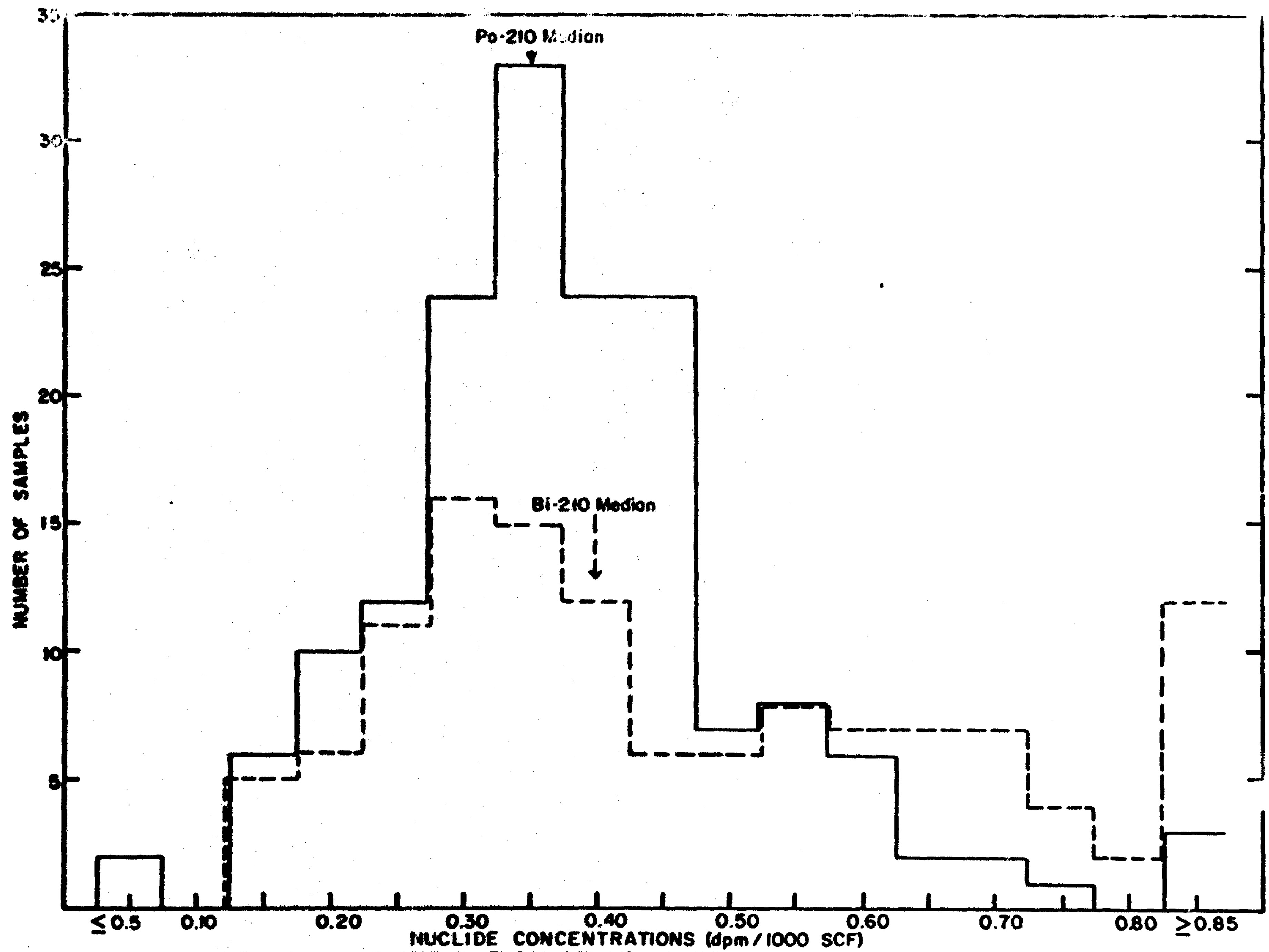


FIGURE 78. FREQUENCY DISTRIBUTION OF MEASURED Bi-210 AND Po-210 CONCENTRATIONS IN HASP SAMPLES

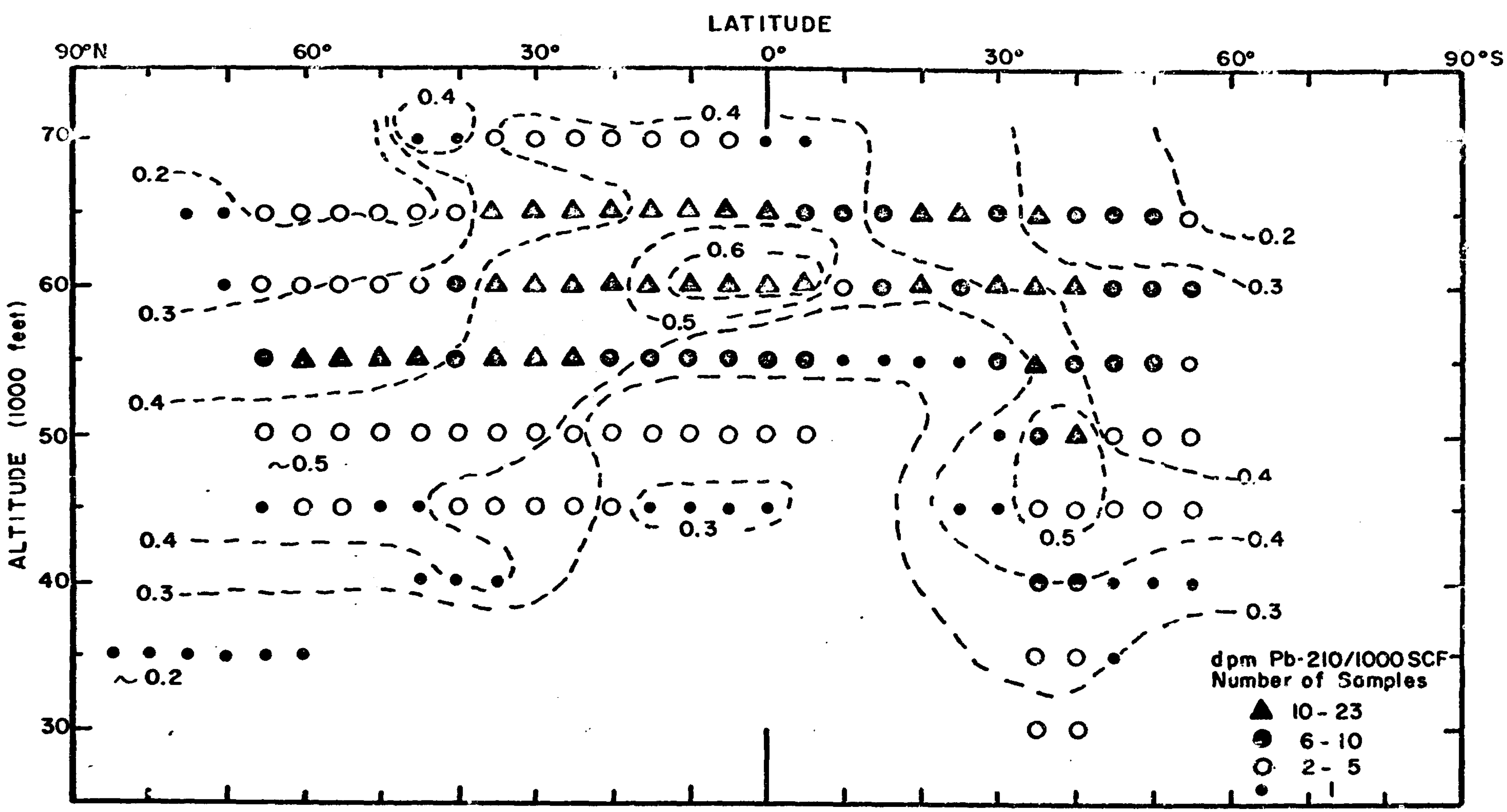


FIGURE 79. THE MEAN DISTRIBUTION OF LEAD-210, OCTOBER 1957 - JULY 1959

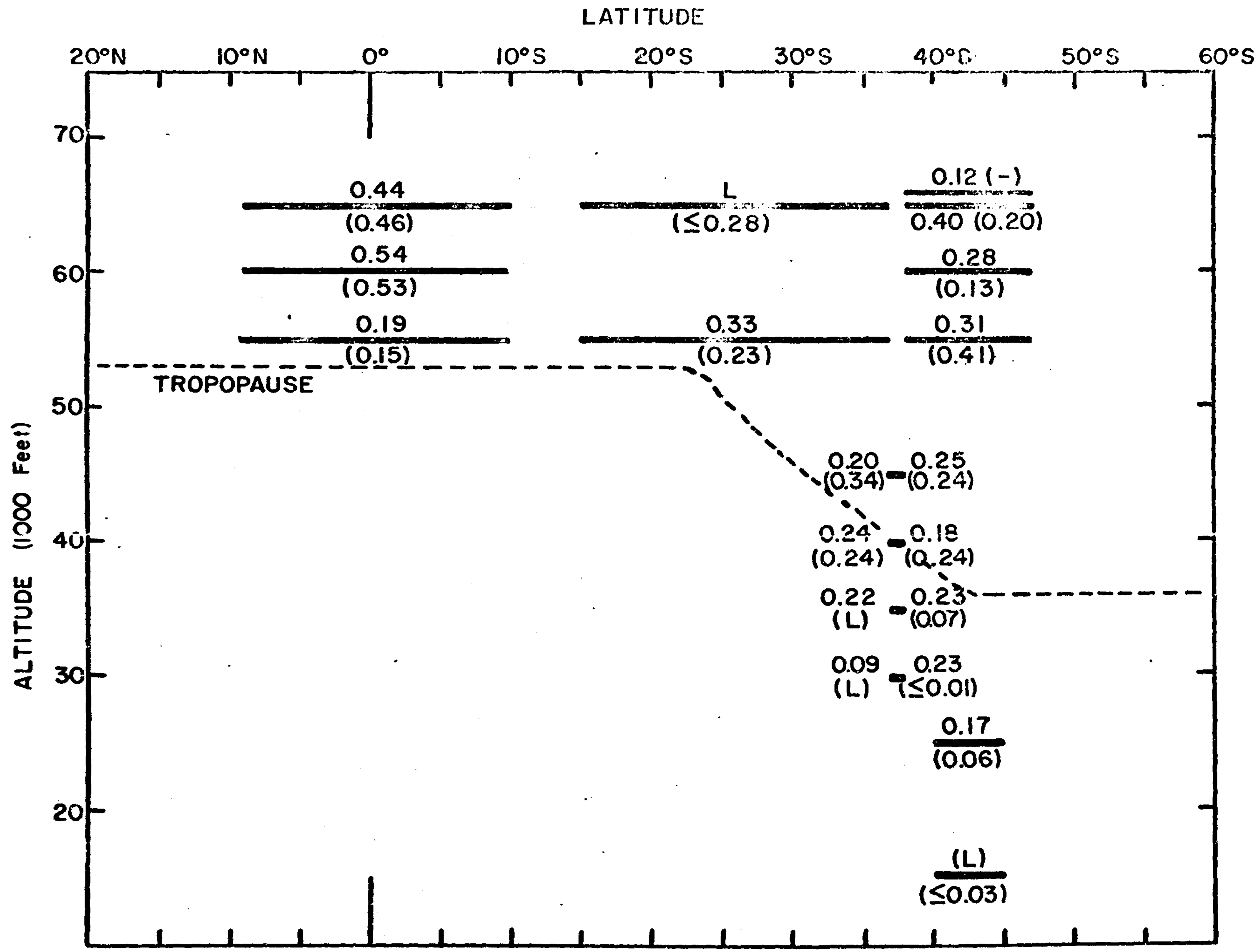


FIGURE 80. MEASUREMENTS (dpm/1000 SCF) OF LEAD-210 AND POLONIUM-210 (IN PARENTHESES) IN DECEMBER 1964 SAMPLES

CHAPTER 7. PROGRESS IN THE DESIGN OF THE STARDUST NUMERICAL MODEL OF TRANSFER AND RAINOUT OF STRATOSPHERIC RADIOACTIVE MATERIALS

Summary of Previous Work

An attempt has been made during Project Stardust to devise a mathematical model of the transfer and rainout of radioactive materials initially introduced into the stratosphere which is capable of reproducing all the known details of the tungsten-185 experiments[20] as well as the general features accompanying polar and tropical stratospheric injections of radioactive material. Because the tungsten-185 tracer experiments have yielded the most useful experimental data, attention has been concentrated on reproducing the prominent features of these data as summarized in DASA-1300, the final report on Project HASP. These features include the meridional distribution of rainout, the time variation of the meridional concentration gradient of tungsten-185 in the stratosphere, the decay of the maximum concentration with time, and the meridional distribution of the height of the maximum concentration in any vertical section. It is desired to devise the simplest model which has the capability of reproducing these observed data. Details of a pure diffusion-rainout model, considered to be the simplest concept, have been given in DASA-1305 and DASA-1309, the Fifth and Ninth Quarterly Reports on Project Stardust. The diffusion process in these models is considered to be the following:

Given a set of axes, the flux of material in the two cardinal directions can be represented as

$$(1) \qquad F_1 = -\rho K_1 \frac{\partial q}{\partial x_1} \qquad \text{and} \qquad F_2 = -\rho K_2 \frac{\partial q}{\partial x_2} \; ,$$

where:

X_1 and X_2 = diffusion axes;

K_1 and K_2 = diffusion coefficients in the X_1 and X_2 directions, respectively ($cm^2\ sec^{-1}$);

ρ = air density ($gm\ cm^{-3}$);

q = mixing ratio of radioactive material.

In global diffusion, the atmospheric horizontal scale is much greater than the vertical scale and, hence, K_1 cannot equal K_2. For this case equations (1) can be correct only if the X_1 and X_2 axes represent the principal axes of the diffusion tensor. The initial models assumed that the principal axes were in the radial direction from the center of the earth, i.e. along the direction of the gravity vector, and in the orthogonal poleward direction.

Computations resulting from this diffusion-rainout model indicated that three of the four important properties of the observed data could be reproduced by this simple model; the property which could not be achieved was the decrease, in the poleward direction, of some 20 thousand feet in the level of the maximum concentration. The early models indicated that no matter what the assumed spatial distribution of diffusion coefficients, the combination of a pure diffusion process in the stratosphere and a rainout-diffusion process in the troposphere will raise, not lower, the level of the maximum concentration in the stratosphere.

At this point numerical experiments commenced incorporating the effects of hypothetical meridional circulations in the diffusion-rainout model. Worthy of note was a seasonally varied "Brewer-Dobson" type meridional circulation suggested by Dr. L. Machta of the U. S. Weather Bureau. Seasonal variation was accomplished by the six months alternate use of circulations applicable to the Northern Hemisphere winter and to the Southern Hemisphere winter (Figures 81a and 81b).

The particular values of the variables used with this circulation yielded results which were incompatible with the observed tracer distributions, but the stratospheric distributions predicted by this model were encouraging in one respect. For the first time a lower elevation of the level of maximum concentration in the polar region than in the equatorial region could be maintained in spite of the tropospheric rainout mechanism. Perhaps some version of this circulation model could ultimately give satisfactory results.

Experimentation was also conducted with some meridional circulation schemes confined to the lower stratosphere in the vicinity of the Northern Hemisphere tropopause gap. The use of this type of circulation was based on the observed movement of debris associated with both tropical and polar injections. With respect to a tropical injection, the transport has been observed to be downward and poleward, whereas, with respect to a polar injection, the transport has appeared to be upward and equatorward in the vicinity of the gap. Although the distribution patterns of concentration were initially encouraging in these experiments, attempts to fit the observed behavior of polar and tropical injections, in detail, with a single circulation pattern, resulted in so many arbitrary modifications that this approach was tentatively abandoned.

It was decided to continue with a rather more complicated, pure diffusion-rainout model. It was felt that the possibilities of this form of the model had not yet been exhausted. Also, a revised estimate of the initial source configuration (first made known to us by the U. S. Weather Bureau) caused some uneasiness in the interpretation of our early experiments which assumed a point source at 70 thousand feet. This revised estimate of the initial source configuration indicates a vertical line source with the center of gravity at 57.5 thousand feet or slightly below. This was an important revision and implied an additional objective of the numerical experiments, namely to simulate a process

which could cause a rise in the level of maximum concentration at the equator from 57.5 thousand feet to 70 thousand feet in about six to twelve months from the time of injection.

In what follows we will summarize briefly the developments during the present reporting period, covering the following topics: (a) development of a general anisotropic diffusion model, (b) studies assuming isentropic surfaces to be general axes of the diffusion tensor, and (c) studies of diffusion and rainout patterns associated with principal diffusion axes arbitrarily selected to be parallel to the observed stratospheric distributions of concentration of material.

General Anisotropic Diffusion Model

A simple generalization of the flux relationship, equation (1), yields

$$F_1 = -\rho\left[K_{11}\frac{\partial q}{\partial X_1} + K_{12}\frac{\partial q}{\partial X_2}\right] , \tag{2}$$

$$F_2 = -\rho\left[K_{21}\frac{\partial q}{\partial X_1} + K_{22}\frac{\partial q}{\partial X_{22}}\right] .$$

The flux expression in equation (2) reduces to the flux expression in equation (1) if the axes used for computation are indeed the principal axes of the diffusion tensor. In our numerical model we are forced to adopt some quasi-cartesian coordinate system for computational purposes. This coordinate system may or may not be the principal axes at a given point. It seems most practical to maintain the quasi-cartesian system for computation and to rotate the diffusion tensor into the numerical computing grid. This can be done if the principal axes and the values of the diffusion coefficients along the principal axes are assumed to be known. For example, if the principal diffusion axes intersect the computational axes at a given point in the numerical grid with an angle α, and if the diffusion coefficients in the principal axis system (K'_{11}, K'_{22}) are assumed known, then the diffusion

coefficients at the grid point in the computational coordinate system are given approximately by

$$
(3)\quad
\begin{aligned}
K_{11} &= \cos^2 \alpha\, K'_{11} + \sin^2 \alpha\, K'_{22} \,, \\
K_{22} &= \sin^2 \alpha\, K'_{11} + \cos^2 \alpha\, K'_{22} \,, \\
K_{12} &= \cos \alpha \cdot \sin \alpha\, K'_{11} - \sin \alpha \cdot \cos \alpha\, K'_{22} \,, \\
K_{21} &= \sin \alpha \cdot \cos \alpha\, K'_{11} - \cos \alpha \cdot \sin \alpha\, K'_{22} \,.
\end{aligned}
$$

Since, in application, α is a very small angle, it is permissible to regard α as the local slope of the principal axis of diffusion. For our geometry α is the angle between the principal diffusion axis and the r, $r_0\,\phi$ (r_0 = the mean radius of the earth and ϕ = latitude) coordinate system which is assumed to be cartesian enough to warrant the transformation of equation (3). For actual computation we use the r, $r_0\,\mu$ system ($\mu = \sin\phi$). The slope in the r, $r_0\,\phi$ system $\left(\frac{\partial Z}{r_0 \partial \phi}\right)$ is related to the slope in the r, $r_0\,\mu$ system $\left(\frac{\partial Z}{r_0 \partial \mu}\right)$ by

$$\frac{\partial Z}{r_0\, \partial \phi} = \frac{\partial Z}{r_0 \partial \mu} \cdot \frac{\partial \mu}{\partial \phi} = \cos\phi \cdot \frac{\partial Z}{r_0 \partial \mu}$$

If we now take the divergence of the flux, equation (2) in the r, $r_0\,\mu$ system, we obtain

$$\frac{\partial q}{\partial t} = \frac{1}{\rho}\left[\frac{\partial}{\partial r}\left(\rho K_{22} \frac{\partial q}{\partial r}\right) + \frac{\partial}{r_0 \partial \mu}\left((1-\mu^2)\rho K_{11} \frac{\partial q}{r_0 \partial \mu}\right)\right.$$

$$\left. + \frac{\partial}{\partial r}\left(\rho \cos\phi\, K_{12} \frac{\partial q}{r_0 \partial \phi}\right) + \frac{\partial}{r_0 \partial \mu}\left(\rho \cos\phi\, K_{21} \frac{\partial q}{\partial r}\right)\right] .$$

Because approximations are involved in the transformations when actually used and because the slopes as well as the $r_0 \cdot \mu$ distances vary from grid point to grid point, it was considered desirable to check the validity of these assumptions. It was assumed that the principal axis of the diffusion tensor had a constant slope of 9.19×10^{-4} with the r, $r_0\mu$ coordinate system, and that $K'_{11} = 3 \times 10^9$ cm^2 sec^{-1}, $K'_{22} = 10^4$ cm^2 sec^{-1} in the principal axis system. The transformations, equations (3), were used to obtain the values of K_{11}, K_{22} and $K_{12} = K_{21}$ in our r, $r_0\mu$ coordinate system. The first two terms on the right in the differential equation (4) were numerically integrated by methods outlined in DASA-1305, the Fifth Progress Report on Project Stardust. The finite difference analogues of the last two terms of equation (4) were treated as centered differences and were included in the general finite difference scheme as explicit variables. These additional terms in finite difference form are:

$$\frac{1}{2\Delta r \cdot r_0 (\mu_{j+1} - \mu_{j-1})} \Bigg\{ \Bigg[\frac{\rho_{j,k+1}}{\rho_{j,k}} (\cos\phi K_{12})_{j,k+1} (q_{j+1,k+1} - q_{j-1,k+1})$$

$$- \frac{\rho_{j,k-1}}{\rho_{j,k}} (\cos\phi K_{12})_{j,k-1} (q_{j+1,k-1} - q_{j-1,k-1}) \Bigg]$$

$$+ \Big[(\cos\phi K_{21})_{j+1,k} (q_{j+1,k+1} - q_{j+1,k-1}) - (\cos\phi K_{21})_{j-1,k} (q_{j-1,k+1} - q_{j-1,k-1}) \Big] \Bigg\}.$$

To avoid boundary effects it was assumed that the source was located at the equator at a height of 21 km. A measure of the adequacy of equations (3) and (4) is the degree to which the level of maximum concentration, as determined from the computations, follows the assumed slope of the principal axis. The extent of the agreement 24 weeks after injection is shown in Figure 82. The level of maximum concentration in a vertical section was estimated by parabolic interpolation of the computed concentration at grid points bracketing the maximum and is consequently subject to some uncertainty. The agreement is considered to be satisfactory and it is worth noting that the unequal horizontal grid increments in the Northern and Southern Hemisphere do not seriously distort the results.

Isentropic Diffusion

The anisotropic diffusion sketched in the preceding section requires that the principal diffusion axis be specified in advance. On physical grounds it is perhaps reasonable to assume that large scale mixing motions, at least in the stratosphere, should occur along isentropic surfaces. The bulk of the experiments carried out during this reporting period were concerned with this hypothesis.

It has been pointed out by several authors that the observed slope of isolines of concentration of radioactive debris in the stratosphere is about twice the observed slope of the relevant isentropic surfaces. For example, at source levels the isentropic surfaces descend about 10 thousand feet from the equator to the North Pole in the winter season and only 5 thousand feet in the summer season. The observed level of maximum concentration of the radioactive debris decreases in height by about 20 thousand feet from the equator to the pole. Clearly, then, the isentropic hypothesis cannot by itself reproduce the desired features of distribution in the stratosphere.

It is well at this point to remark on the nature of the observations which lead to the conclusion that the observed level of maximum concentration decreases by some 20 thousand feet from the equator to the pole. In particular, one must inquire how soon after injection this statement is true. If the center of gravity of the initial injection was at 70 thousand feet, then the statement is true within a very few months after injection. If, however, the center of gravity of the initial injection was at 60 thousand feet or below, then the statement is not true for at least the first six months after injection. Going back to the original data tabulation, i.e. Table 4.3 in DASA-1300, the final report on Project HASP, the flight paths in the mid-latitude stratosphere clearly show that the level of maximum concentration at a fixed time is about 55 thousand feet or below within a few months after injection. Unfortunately, there is really no firm evidence within the first six to nine months after injection as to where the level of maximum concentration is at the source latitude. The evidence is compelling, however, that after a period of about one year, the maximum concentration at the source latitude is in the neighborhood of 70 thousand feet.

Figure 83 schematically illustrates a possible time development of the level of maximum concentration as a function of latitude under the hypothesis that the major injection was at about 57.5 thousand feet. It is to be noted that after three months the slope of the observed distribution is approximately that of the isentropic surface at that level. As time progresses the slope of the isentropic surface is exceeded.

What process, then, can contribute to the slow rise of the source in equatorial regions? Clearly, only general circulation terms of the type suggested by Dr. Machta (Figure 1) or a very pronounced height dependency of the horizontal diffusion coefficient such that there is a very large lateral mixing from 50 to 60 thousand feet and relatively small lateral mixing above 60 thousand feet.

The latter hypothesis was investigated quite intensively during the present reporting period. In brief, varying degrees of negative results were obtained in all cases. By suitable height variation of the lateral diffusion coefficients, the height of the maximum concentration can be caused to rise 10 to 15 thousand feet in one year. A variation by a factor of 5 or 10 in the lateral diffusion coefficient is necessary to accomplish this. However, the other features of the observed tungsten-185 distribution and fallout were difficult to reproduce with a pure diffusion model.

The most elaborate experiment in this series involved variation of the slopes of the isentropic surfaces seasonally, to set the vertical diffusion coefficient in the gap region proportional to the magnitude of the monthly mean jet and to fix the horizontal diffusion coefficients according to the vertical and meridional distribution of the vector standard deviation of the wind (extrapolating values into the stratosphere).

Data for these seasonal and spatial variations were obtained from U.S. Weather Bureau published cross-sections[21,22]. In order to cause the source to rise at the latitude of injection it was necessary to hypothesize quite large horizontal diffusion coefficients at 55 and 60 thousand feet compared to those assumed to hold at 65 and 70 thousand feet in the tropical region.

The results were not satisfactory. The large diffusion coefficients in the gap region and the strong tropospheric rainout mechanism made it impossible to maintain the desired slope of the level of maximum concentration in the stratosphere equatorward of the gap region, although more satisfactory results were obtained poleward of the gap region. In order to remedy this situation the following strategems were tried:

(1) rainout was effected every sixth complete computation cycle, rather than every cycle, and

(2) particle terminal velocity was varied as a function of latitude according to the latitudinal and seasonal variation of stratospheric density as given by Cole and Kantor[23].

Neither of these alternatives substantially altered the results. Our conclusion is that, if the initial injection is assumed to be at 55 or 60 thousand feet, it is almost impossible to reproduce the observed features with a pure diffusion model.

Anisotropic Diffusion Model with Arbitrary Slope of the Diffusion Axis

A few experiments were run with the following joint hypothesis:

(1) the principal diffusion axis is identical with the slope of the level of maximum concentration as observed one year after injection, and

(2) the center of gravity of the initial injection is at 70 thousand feet.

The preliminary results with this model were gratifying. The desired stratospheric slope of the level of maximum concentration was maintained even after substantial rainout was effected. Also, a mid-latitude maximum of rainout was obtained without hypothesizing a gap region. This is due to a combination of the effect of the hypothesized slope of the vertical diffusion coefficient and the meridional distribution of the removal factor, which is proportional to the mean annual rainfall.

A north polar injection tested using the same model also produced gratifying results in that the characteristic slope upwards toward the equator was maintained even after substantial rainout. In short, it is felt that the significant features of the tungsten-185 data and of the history of changes in the distribution of Soviet debris in the stratosphere can be reproduced by this model, provided, of course, that the joint hypothesis stated at the beginning of this section can be justified. In a way, the first part of this hypothesis

already assumes the result. There is some evidence in a recent unpublished paper by Hering and Borden[24] that the slope of the diffusion axis assumed in this experiment is roughly the same as the slope of the lines of equal potential vorticity in the stratosphere. From a physical point of view it is not obvious that lines of constant potential vorticity should be a principal axis of the diffusion tensor. The sensitivity of the results to the second part of the hypothesis, namely that the injection was at 70 thousand feet, has not yet been examined.

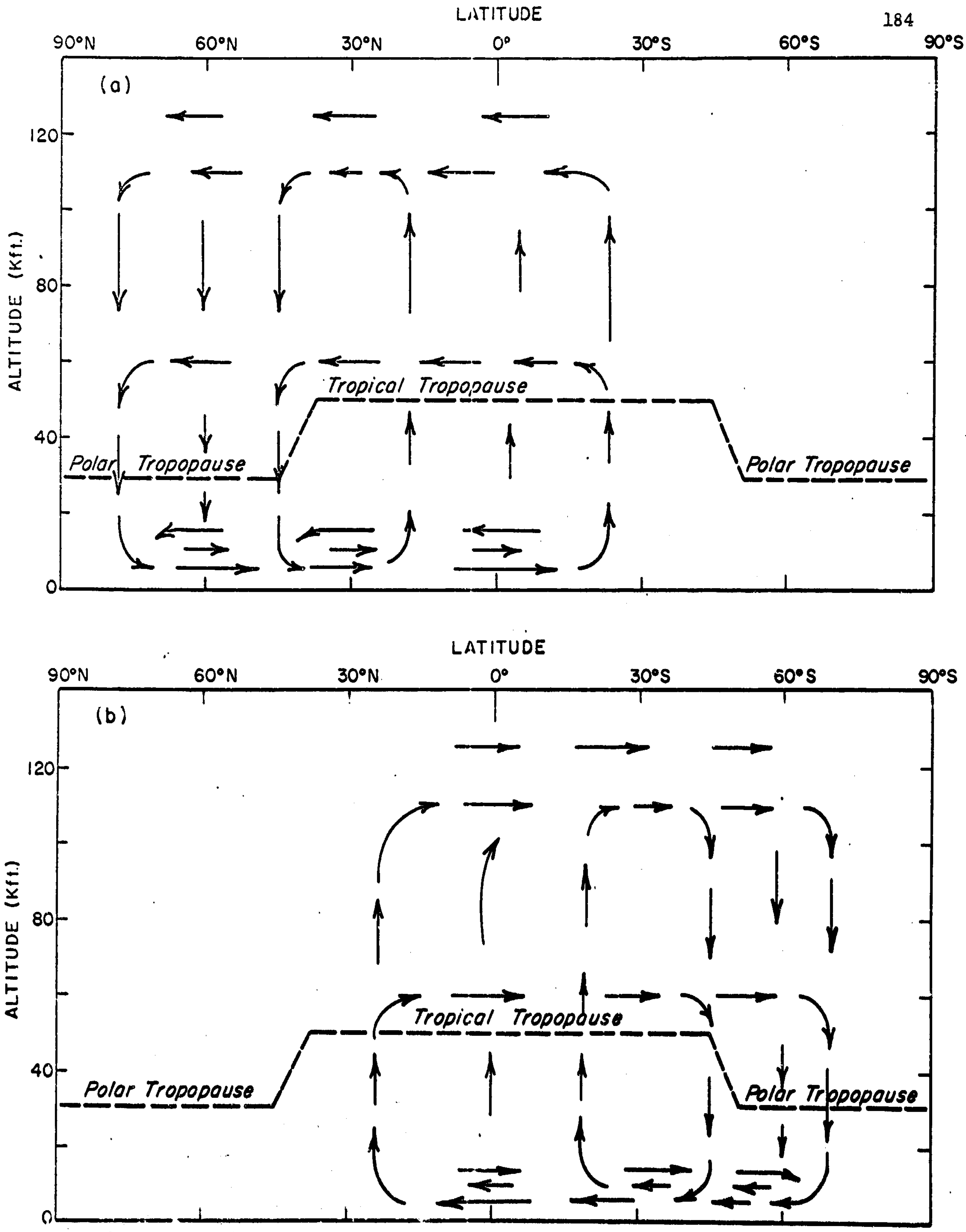


Fig. 31. Schematic Representation of Meridional Circulations (Machta) for N. Hemisphere Winter (a) and S. Hemisphere Winter (b).

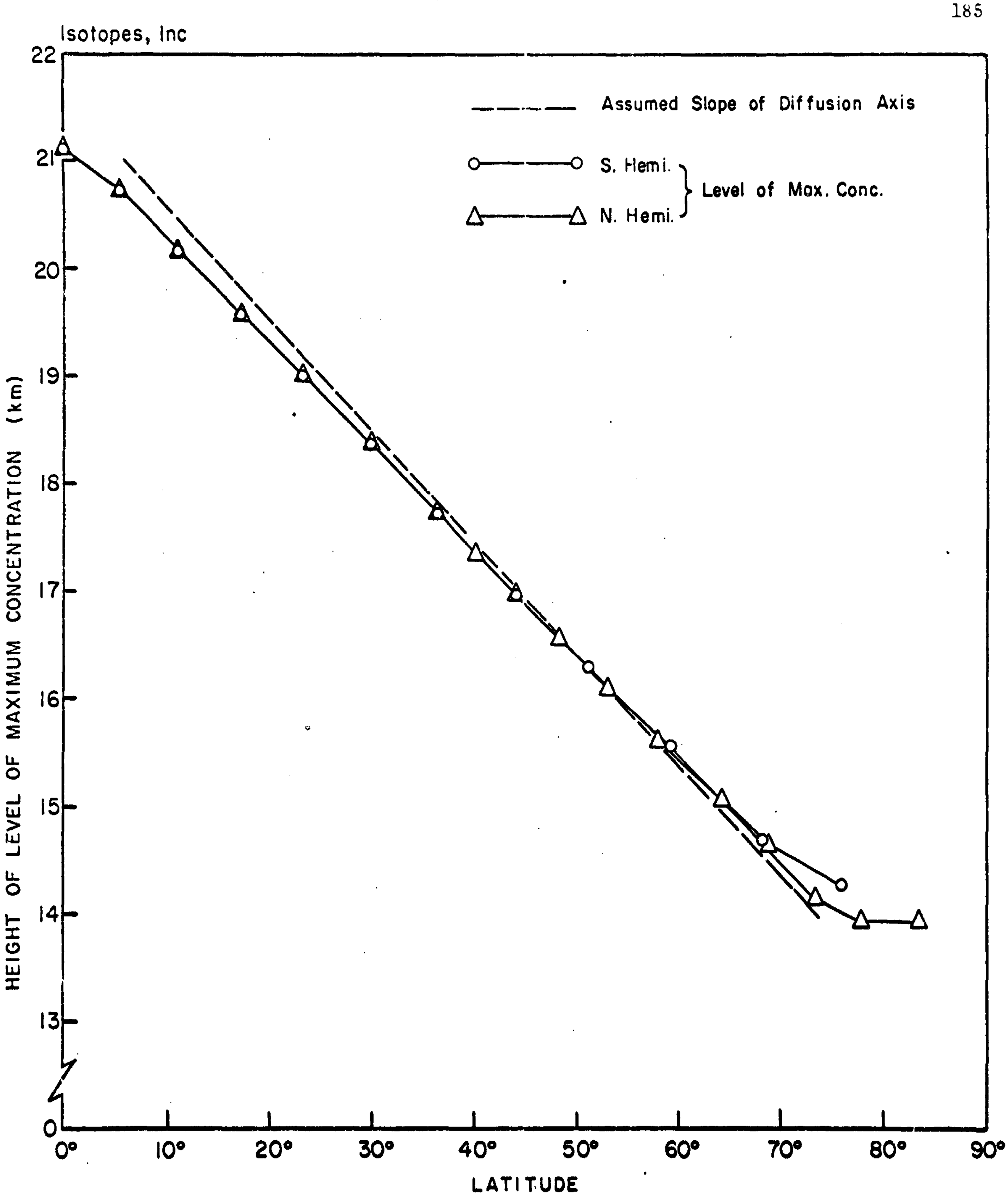


FIGURE 82. VARIATION WITH LATITUDE OF THE LEVEL OF MAXIMUM CONCENTRATION PREDICTED BY THE NUMERICAL MODEL 6 MONTHS AFTER INJECTION COMPARED WITH ASSUMED SLOPE OF THE PRINCIPAL DIFFUSION AXIS

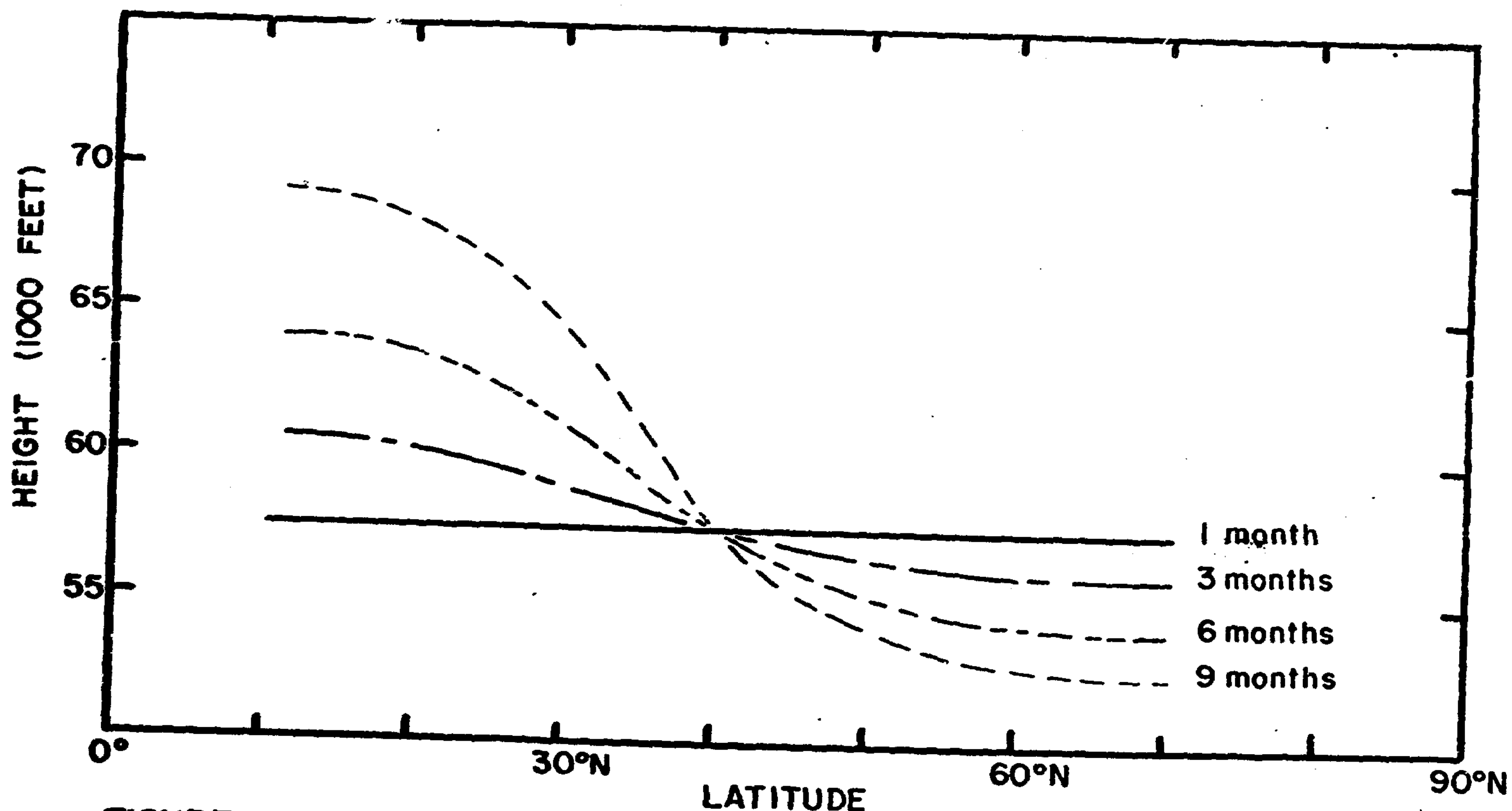


FIGURE 83. SCHEMATIC REPRESENTATION OF POSSIBLE TIME DEVELOPMENT OF LEVEL OF MAXIMUM CONCENTRATION ASSUMING MAJOR INJECTION WAS AT 57,500 FEET

CHAPTER 8. THE CARBON-14 CONCENTRATION IN GROUND-LEVEL AIR

The measurement of the concentration of carbon-14 in the carbon dioxide of ground-level air in northern New Jersey was begun in January, 1960 during Project HASP, and has continued during Project Stardust. Currently one sample is collected each month at the Township of Washington, Bergen County, New Jersey.

The samples are collected by exposing to the atmosphere approximately 200 ml. of 4N KOH solution contained in two Petri dishes. Approximately once per week the Petri dishes are emptied into a polyethylene bottle and are then refilled with fresh KOH solution. On about the first day of each month the accumulated sample, consisting of about 800 ml., is taken to the laboratory and is processed. The solution is acidified with phosphoric acid to release the dissolved carbon dioxide. The evolved gas is collected and is purified twice using calcium oxide furnaces. It is then admitted to a two liter proportional counter and is counted twice, the first time for approximately 300 minutes and the second time for approximately 900 minutes.

The results of measurements of samples collected during September, 1963 to December, 1965 are listed in Table 13. They are expressed in percent of activity above the activity of the National Bureau of Standards oxalic acid carbon-14 standard. They are plotted together with results of measurements of earlier samples (reported in DASA-1306 and DASA-1309) in Figure 84.

During 1960 and 1961 concentrations ranged between 10 and 25 percent above the NBS standard. In 1962, as carbon-14 produced by the 1961 Soviet weapons tests series began to reach the troposphere, carbon-14 concentrations rose, reaching a peak value of 42 percent above the NBS standard in September, 1962. In late 1962 concentrations fell, as they had in late 1960 and late 1961, and as they would again in late 1963, late 1964 and late 1965.

By April, 1963 large quantities of carbon-14 from the 1961 and 1962 weapons tests series had begun to enter the troposphere, and the carbon-14 concentrations rose rapidly, reaching a value of 91 percent above the NBS standard in August, 1963. After decreasing during the winter of 1963-1964, concentrations again rose to values in excess of 90 percent above the NBS standard in mid-1964. They decreased again during the winter of 1964-1965, and rose only to values 70 to 75 percent above the NBS standard in mid-1965. They decreased again with the onset of the winter of 1965-1966.

TABLE 13

CARBON-14 CONCENTRATIONS IN GROUND-LEVEL AIR AT THE TOWNSHIP OF WASHINGTON, BERGEN COUNTY, NEW JERSEY. (CONCENTRATIONS ARE IN PERCENT ABOVE THE ACTIVITY OF THE NBS OXALIC ACID STANDARD.)

Sample Number	Collection Interval	δ C^{14}
SD-50	31 Aug 63 - 16 Sep 63	+ 90.0
SD-51	16 Sep 63 - 1 Oct 63	+ 85.1
SD-52	1 Oct 63 - 15 Oct 63	+ 75.2
SD-53	15 Oct 63 - 31 Oct 63	+ 75.3
SD-54	31 Oct 63 - 16 Nov 63	+ 78.9
SD-55	16 Nov 63 - 1 Dec 63	+ 65.1
SD-56	1 Dec 63 - 16 Dec 63	+ 86.0
SD-57	16 Dec 63 - 2 Jan 64	+ 77.8
SD-58	3 Jan 64 - 20 Jan 64	+ 60.8
SD-59	20 Jan 64 - 3 Feb 64	+ 75.0
SD-60	3 Feb 64 - 17 Feb 64	+ 75.6
SD-61	17 Feb 64 - 2 Mar 64	+ 71.5
SD-62	2 Mar 64 - 14 Mar 64	+ 71.5
SD-63	14 Mar 64 - 31 Mar 64	+ 78.8
SD-64	31 Mar 64 - 16 Apr 64	+ 75.5
SD-65	16 Apr 64 - 1 May 64	+ 78.5
SD-66	8 May 64 - 15 May 64	+ 86.3
SD-67	15 May 64 - 31 May 64	+ 89.8
SD-68	31 May 64 - 16 Jun 64	+ 91.5
SD-69	16 Jun 64 - 1 Jul 64	+ 78.6
SD-70	1 Jul 64 - 15 Jul 64	+ 91.4
SD-71	15 Jul 64 - 1 Aug 64	+ 92.3
SD-72	1 Aug 64 - 17 Aug 64	+ 92.5
SD-73	17 Aug 64 - 1 Sep 64	+ 83.4
SD-74	1 Sep 64 - 16 Sep 64	+ 90.9
SD-75	16 Sep 64 - 1 Oct 64	+ 88.7
SD-76	1 Oct 64 - 17 Oct 64	+ 85.4
SD-77	17 Oct 64 - 31 Oct 64	+ 80.0
SD-78	31 Oct 64 - 15 Nov 64	+ 73.7
SD-79	15 Nov 64 - 1 Dec 64	+ 76.1
SD-80	1 Dec 64 - 15 Dec 64	+ 64.5
SD-81	15 Dec 64 - 2 Jan 65	+ 64.2
SD-82	2 Jan 65 - 19 Jan 65	+ 67.2
SD-83	19 Jan 65 - 30 Jan 65	+ 68.0
SD-84	30 Jan 65 - 15 Feb 65	+ 69.9
SD-85	15 Feb 65 - 28 Feb 65	+ 69.7
SD-86	28 Feb 65 - 15 Mar 65	+ 70.2
SD-87	15 Mar 65 - 31 Mar 65	+ 72.1
SD-88	31 Mar 65 - 15 Apr 65	+ 74.1
SD-89	15 Apr 65 - 4 May 65	+ 71.7
SD-90	4 May 65 - 21 May 65	+ 72.5

TABLE 13

CARBON-14 CONCENTRATIONS IN GROUND-LEVEL AIR AT THE TOWNSHIP OF WASHINGTON, BERGEN COUNTY, NEW JERSEY. (CONCENTRATIONS ARE IN PERCENT ABOVE THE ACTIVITY OF THE NBS OXALIC ACID STANDARD) - (Continued)

Sample Number	Collection Interval	δC^{14}
SD-91	21 May 65 - 2 Jun 65	+ 72.1
SD-92	2 Jun 65 - 1 Jul 65	+ 74.5
SD-93	1 Jul 65 - 1 Aug 65	+ 73.0
SD-94	1 Aug 65 - 3 Sep 65	+ 72.7
SD-95	3 Sep 65 - 1 Oct 65	+ 67.0
SD-96	1 Oct 65 - 31 Oct 65	+ 68.0
SD-97	31 Oct 65 - 2 Dec 65	+ 71.0
SD-98	2 Dec 65 - 1 Jan 66	+ 57.8

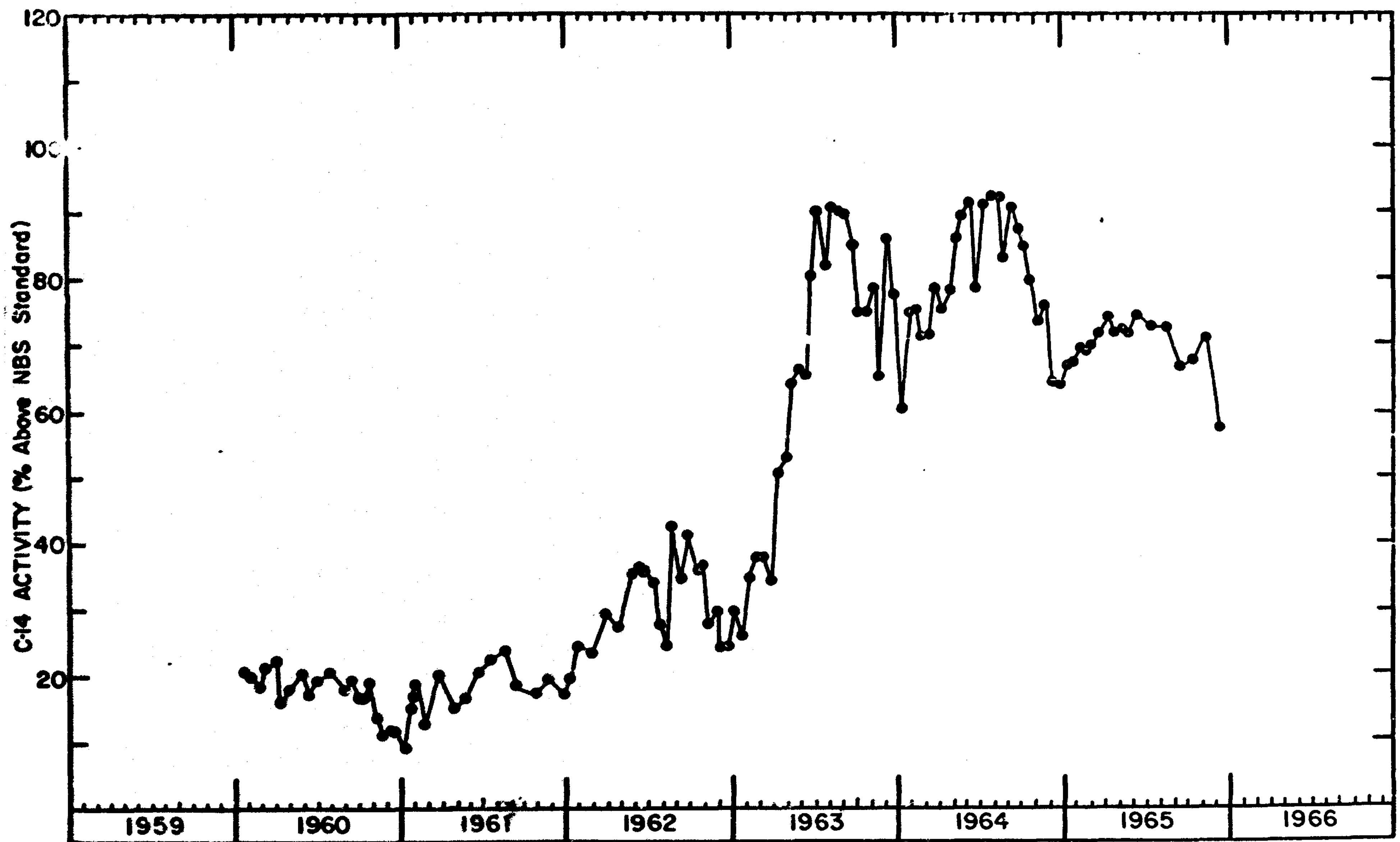


FIGURE 84. ACTIVITY OF ARTIFICIAL CARBON-14 IN GROUND LEVEL AIR, TOWNSHIP OF WASHINGTON, BERGEN COUNTY, NEW JERSEY

CHAPTER 9. THE DISTRIBUTION OF RADIOACTIVE FALLOUT IN TWO NEW JERSEY SOILS, 1960 to 1963

The distribution of the fission products strontium-90, cesium-137, ruthenium-106 and cerium-144 was studied in eleven New Jersey and two Kansas soils and the results were reported in DASA-1300, the Final Report on Project HASP, and by Walton[25]. Subsequently seven additional Kansas soils were analyzed for the same nuclides, and results were reported in DASA-1309, the Ninth Progress Report on Project Stardust. Both of these investigations were designed to determine the concentration and vertical distribution of these fallout nuclides, which constitute a potential health hazard, as a function of such factors as soil composition, structure, local climate and topography. A logical extension of these studies is an investigation of the change in vertical distribution of radioactivity in soil with time. Does the depth of soil which must be sampled in order to obtain approximately 100% of the radioactivity increase with time, and by how much for the different nuclides? How does the shape of the vertical profiles of the activity of the different radionuclides change with time as percolating water carries the radionuclides deeper, and as more fallout is introduced at the surface? How do fallout burdens calculated from measurements of fallout in soil compare with those calculated from data on cumulative fallout measured in rainfall?

Sampling and Analytical Procedures

Two New Jersey soils were chosen for repeated sampling and analysis of four fission products in an attempt to answer some of these questions. The soils, Marlton sandy loam (sample 8) and Lakewood sand (sample 10), were first sampled for the HASP study on 8 August 1960. They were chosen for the extended study because of their proximity to each other, the differences in their soil characteristics, and the difference in distribution of radioactivity with depth they showed in the HASP

study. The soils were resampled on 11 January 1962 (samples 8A and 10A), 11 October 1962 (samples 8B and 10B) and on 25 September 1963 (samples 8C and 10C). All soil cores were taken as close as possible to the original coring locations utilizing the same coring device used for the earlier soil studies.

The procedures used for sample collection, processing and analysis were the same as those described in DASA-1300 and DASA-1309. The cores collected were 10 or 12 inches in diameter. To minimize vertical contamination that might have occurred during sample collection, an 8 inch core was taken from the center of the 10 or 12 inch core. The 8 inch core was cut into layers, each of which was taken as a separate sample. Depths of each of the layers below the top of the core were, in general, the same in all cores: 0 to 1/2 inch, 1/2 to 1 inch, 1 to 2 inches, 2 to 3 inches, 3 to 4 inches, 4 to 6 inches, 6 to 9 inches, 9 to 12 inches, 12 to 18 inches and, in several cases, a sample below 18 inches of varying thickness. Thinner samples were taken from the upper layers because these layers were expected to contain the highest concentrations of radioactivity. These samples were dried, crushed, ground and homogenized.

In general, the physical characteristics (color, texture, amount of organic matter) of each of the sample layers in each of the soils were similar from year to year. However, in one or two instances the samples from the upper layers of the cores collected in different years, which supposedly represent the same soil horizon, are markedly different in the amount of organic matter they contain.

To check the reliability of the results of the analyses of the New Jersey soil samples, a series of "blank samples" of soil collected before 1945, and thus containing no fallout radioactivity, was measured at the same time as the regular soil samples. Any activity found in these "blank samples" must be attributed to contamination picked up during the analyses. Equivalent contamination of the regular samples is to be expected. In so far as possible the section of the laboratory in which the soil analyses were being performed was isolated from the remainder of the

laboratory in which other types of samples containing fission products were being analyzed. New glassware and fresh reagents were used in the soil analyses. Nevertheless the "blank samples" analyzed with samples 8A, 8B, 8C, 10A, 10B and 10C displayed significant activities of each of the nuclides analyzed.

The results for the blank samples analyzed with the New Jersey soil samples are summarized in Table 14. Some analyses of samples 8A, 10A, 8B and 10B were performed during 1962 and 1963, and data for the first group of 14 blank samples are relevant to them. Other analyses of samples 8A, 10A, 8B, and 10B were performed during 1964, as were all analyses of samples 8C and 10C, and data for "blank samples" B and C are relevant to them. Apparently the "blank samples" analyzed in 1964 were no less subject to contamination than were those analyzed in 1962 and 1963.

Results

The weights of the various intervals sampled in each core of the Marlton sandy loam are listed in Table 15, and those for the Lakewood sand are listed in Table 16. The results of the radiochemical analysis of the cores of the Marlton are given in Table 17, and those for the Lakewood are given in Table 18. The data from the original study of these soils under HASP (samples 8 and 10) are included in Tables 15 to 18 for comparison with the new data. Analytical results for each nuclide are given in disintegrations per minute per 100 grams of soil (dpm/100 g) in Tables 17 and 18. Calculating results on a per-unit-weight basis and using weight of overlying soil as a measure of depth permits comparison of the different soils and cores at various levels without regard to possible compaction of the soil during the coring or sampling operations. In addition, for each of the intervals sampled the total activity in that interval and the cumulative activity from the surface to the bottom of that interval are given. The sampling intervals are listed in inches of depth, and in Tables 15 and 16 the weight of soil (in grams) in each interval, the cumulative weight to the bottom of each interval, and the weight of soil above the

midpoint of each interval are given.

Comparison of the data in Table 14 with those in Tables 17 and 18 suggests that the activities of fission products in most samples taken from below a depth of two inches in the cores were similar to the activities found in the "blank samples", so that all such activities must be considered of doubtful reliability.

The Vertical Distribution of Fallout Radionuclides

The Marlton sand loam and Lakewood sand were chosen for this study because of their differences in soil characteristics and because, in the orginal study under Project HASP, they each demonstrated a vertical profile of radioactivity which was both internally consistent for each nuclide, relatively consistent in comparisons between nuclides, and which was reasonable in reference to the physical and chemical characteristics of each soil. The results of the present study do not always show the same degree of consistency.

Figures 85 to 88 have been prepared to facilitate comparison of the vertical profiles of the nuclides in the different cores. Strontium-90 and cesium-137 data are plotted in Figures 85 and 86 for the Marlton and Lakewood, respectively. Cerium-144 and ruthenium-106 data are plotted in Figures 87 and 88 for the Marlton and Lakewood, respectively. In the figures the ratios of the total activity above each level to the total activity above the 12 inch level have been plotted against the ratios of total weight of soil above each level to the total weight of soil above the 12 inch level. This eliminates differences in shape between profiles for different cores which result from some cores containing more activity than others. Such differences are discernible in the tables of radiochemical data, Tables 17 and 18. The differences in shape which remain are those which result from differences in distribution of the activity between the upper and lower sections of the cores.

The characteristics of the plots in Figures 85 to 88 is such that a straight line inclined 45° to the abscissa will result if the nuclide is distributed uniformly with depth (for example, ruthenium-106 in Cores 10B and 10C: right hand

side of Figure 88), and a curve convex toward the upper left will result if the nuclide is concentrated preferentially in the uppermost layers of the core (for example, cesium-137 in the Lakewood: right hand side of Figure 86).

Cesium-137 shows variations with depth which change less from one curve to the next than do the variations shown by the other nuclides studied. The scatter in the data, however, do not permit a definitive conclusion as to exact changes with time in the vertical profiles even of cesium-137 during 1960 to 1963. There is some indication (Tables 17 and 18) that the cesium-137 profiles for the samples taken in January 1962 (cores 8A and 10A) are somewhat flatter than those for the soils collected in July 1960 (cores 8 and 10), indicating downward migration of cesium-137. The fact that the vertical profiles, if anything, become steeper for the last two collections (cores 8B, 10B, 8C and 10C) suggests that these last two collections may contain additional fallout from the 1961 and 1962 weapons tests. Perhaps the sampling and analytical precision are such that no significant changes with time in the vertical profile of cesium-137 can be seen below two inches depth in the cores. All profiles of cesium-137 for both soils (Figures 85 and 86) retain essentially the same characteristics as evidenced in the original analysis under Project HASP, with the activity concentrated in the upper inch of the core.

The vertical profiles of strontium-90 display much more scatter in the data than do those of cesium-137, but any changes in the vertical distribution of strontium-90 have not exceeded the uncertainty resulting from variability in the sampling analytical procedures. It is possible, however, that the cores 8A and 8B of the Marlton display the same indication mentioned above for cesium-137, that by 1962 some downward migration of strontium-90 had occurred in the soil column. The strontium-90 profiles of cores 8A and 8B are much flatter than is the profile of core 8.

Data for both cerium-144 and ruthenium-106 are difficult to analyze. These nuclides have much shorter half lives than do strontium-90 and cesium-137

(about 1 year compared to 30 years), and show a correspondingly larger scatter in their vertical profiles in all four cores of both soils. Nevertheless, results of analyses of the January, 1962 collection (cores 8A and 10A) also show a decrease in the vertical gradient of activity compared with cores 8 and 10 (Figures 87 and 88). In general, data for cores 8B, 8C, 10B and 10C indicate that steeper gradients were reestablished by additional fallout during 1962 and 1963. The apparent flattening in the January, 1962 activity profiles probably resulted from downward migration of the fallout nuclides during 1960 to 1962, while the rate of deposition of fallout in the surface was a minimum as a result of the 1958 - 1961 moratorium on testing of nuclear weapons.

Comparison of Measured and Calculated Fallout Deposits

It is of interest to compare the total deposits of fallout nuclides measured in the soil samples with the expected deposits calculated from deposition in precipitation at nearby rain monitoring stations. This comparison will be restricted to strontium-90 and cesium-137. Cerium-144 and ruthenium-106 will not be considered because of the relatively great scatter of data for those nuclides in the soils in this study, and because their much shorter half lives (about 1 year compared to about 30 years for strontium-90 and cesium-137) makes it difficult to estimate the concentrations of these nuclides in fallout, and they were not actually measured in rainfall for much of the duration of this study.

Table 19 summarizes the pertinent data. It gives total strontium-90 and cesium-137 activity, in millicuries per square mile, represented by each core and by cumulative rainfall activity, corrected for decay. The rainfall data are activities measured in rainwater at Westwood, New Jersey, the nearest monitoring station to the soil coring sites. Westwood is, however, almost one hundred miles away from the soil coring sites, a significant distance in terms of rainfall and fallout variability. Indeed, the total rainfall and concentrations of radioactive

fallout at Westwood often differ significantly from those recorded in New York City, less than twenty miles away. Moreover, during the period July 1960 through July 1961, the activity of cesium-137 in rainfall was not actually measured at Westwood, and it has been necessary to calculate it from the measured strontium-90 values by multiplying by the fission production ratio of 1.7.

Comparison of cumulative strontium-90 and cesium-137 activities in the A, B, or C cores with the rainfall data indicates considerable variation of the soil values from the steadily increasing deposit estimated to have fallen out in the New Jersey area. However, when the cumulative activities from both Marlton and Lakewood soils are averaged, the resulting values agree reasonably well with the estimated rainfall deposits and also show a steady increase.

The fact that these cumulative data are closer to expected values when averaged emphasizes an important point in this study; in order to be able to distinguish variations in soil activity over the course of a few years and to be able to understand the causes for the variations, one must know first of all the variation at a single coring-site within the same soil, i.e. a number of cores must be taken from the site covering an area several tens of feet in diameter. Next one must know the short term variations to be expected in soil activity, i.e. fluctuations with change in rainfall, temperature, and level of the water table, so that cores taken years apart may be either taken under exactly the same conditions or so that the differences in these conditions can be accounted for. In addition, the extent of seasonal variations in the soil activity must be known, and must be related to a knowledge of the amount and time distribution of rainfall and of fallout in rainfall at the coring site. Finally, one must have information about changes in the nature of each soil with location and with depth; changes in the organic content, soil minerals, pH, cation exchange capacity, porosity, water content, etc.

TABLE 14

RESULTS OF ANALYSES OF "BLANK SAMPLES" OF SOIL COLLECTED BEFORE 1945

Sample	dpm Sr^{90}	dpm Cs^{137}	dpm Ce^{144}	dpm Ru^{106}
Analyzed during 1962-1963. Activities corrected for decay to 11 January 1962:				
1	-	11.6 ± 1.1	-	-
2	6.0 ± 0.7	16.2 ± 0.9	29 ± 2	23 ± 1.0
4	-	4.1 ± 0.7	≤38	5.5 ± 1.0
5	3.5 ± 2.2	6.5 ± 0.6	22 ± 8	10.4 ± 1.9
6	-	17.6 ± 1.2	90 ± 8	28 ± 2.5
7	3.9 ± 0.8	13.0 ± 1.1	-	36 ± 2.5
8	4.1 ± 1.2	8.6 ± 1.0	68 ± 11	17.9 ± 1.8
9	≤1.7	6.5 ± 1.0	70 ± 7	28 ± 2.6
10	-	-	-	16.7 ± 1.6
11	4.0 ± 0.6	5.7 ± 0.8	82 ± 6	3.6 ± 1.0
12	-	5.5 ± 1.4	69 ± 6	13.3 ± 2.5
13	-	6.5 ± 1.9	77 ± 9	60 ± 6.7
14	-	-	76 ± 13	50 ± 2.9
Mean	3.8 ± 1.4	9.3 ± 4.6	62 ± 24	24 ± 17
Analyzed during 1964. Activities corrected for decay to 11 October 1962:				
B	≤6.8	≤29	192 ± 5	-
Analyzed during 1964. Activities corrected for decay to 25 September 1963:				
C	9.8 ± 6.6	≤14	30 ± 3	≤28

TABLE 15

WEIGHT OF SOIL IN SAMPLED INTERVALS OF CORES OF MARLTON SANDY LOAM

Depth Interval (inches)	Dry Weight of Soil in Interval (grams)	Total Weight Above Bottom of Interval (grams)	Total Weight Above Mid-Point of Interval (grams)
Marlton-8, Collected 8 Aug 1960			
0 - 1/2	220	220	110
1/2 - 1	409	629	424
1 - 2	960	1,589	1,109
2 - 3	1,178	2,767	2,178
3 - 4	1,418	4,185	3,476
4 - 6	2,468	6,653	5,419
6 - 10	5,248	11,901	9,927
10 - 12	2,624	14,525	13,213
Marlton-8A, Collected 11 Jan 1962			
0 - 1	1,135	1,135	568
1 - 2	908	2,043	1,589
2 - 3	1,022	3,064	2,554
3 - 6	3,064	6,129	4,597
6 - 9	3,178	9,307	7,718
9 - 12	3,178	12,485	10,896
12 - 18	6,924	19,408	15,947
18 - 21 1/2	3,292	22,700	21,054
Marlton-8B, Collected 11 Oct 1962			
0 - 1	473	473	236
1 - 2	1,050	1,523	998
2 - 3	1,044	2,567	2,045
3 - 6	4,277	6,844	4,706
6 - 9	4,766	11,610	9,227
9 - 12	3,458	15,068	13,339
12 - 18	7,619	22,687	18,878
18 - 23 1/2	6,875	29,562	26,124
Marlton-8C, Collected 25 Sep 1963			
0 - 1/2	539	539	270
1/2 - 1	702	1,241	890
1 - 2	1,027	2,268	1,754
2 - 3	1,264	3,532	2,900
3 - 4	1,273	4,805	4,168
4 - 6	2,253	7,058	5,932
6 - 9	3,440	10,498	8,778
9 - 12	3,707	14,205	12,352
12 - 18	7,721	21,926	18,066

TABLE 16

WEIGHT OF SOIL IN SAMPLED INTERVALS OF CORES OF LAKEWOOD SAND

Depth Interval (inches)	Dry Weight of Soil in Interval (grams)	Total Weight Above Bottom of Interval (grams)	Total Weight Above Mid-Point of Interval (grams)
Lakewood-10, Collected 8 Aug 1960			
0 - 1/2	409	409	204
1/2 - 1	632	1,041	725
1 - 2	1,123	2,164	1,602
2 - 3	1,251	3,415	2,790
3 - 4	1,240	4,655	4,035
4 - 6	2,713	7,368	6,012
6 - 9	4,003	11,371	9,370
9 - 12	4,003	15,374	13,372
Lakewood-10A, Collected 11 Jan 1962			
0 - 1	1,135	1,135	568
1 - 2	1,362	2,497	1,816
2 - 3	1,362	3,859	3,178
3 - 6	3,859	7,718	5,788
6 - 9	4,313	12,031	9,874
9 - 12	3,972	16,003	14,017
12 - 18	7,037	23,040	19,522
Lakewood-10B, Collected 11 Oct 1962			
0 - 1	1,035	1,035	518
1 - 2	1,124	2,159	1,597
2 - 3	995	3,154	2,656
3 - 6	3,320	6,474	4,814
6 - 9	4,015	10,489	8,482
9 - 12	4,240	14,729	12,609
12 - 18	7,810	22,539	18,634
18 - 23 1/2	6,360	28,899	25,719
Lakewood 10-C, Collected 25 Sep 1963			
0 - 1/2	928	928	464
1/2 - 1	652	1,580	1,254
1 - 2	1,195	2,775	2,178
2 - 3	1,243	4,018	3,396
3 - 4	1,500	5,518	4,768
4 - 6	2,588	8,106	6,812
6 - 9	4,116	12,222	10,164
9 - 12	3,783	16,005	14,114
12 - 16	5,142	21,147	18,576
16 - 20	4,900	26,047	23,597

TABLE 17

RESULTS OF RADIOCHEMICAL ANALYSES OF MARLTON SANDY LOAM

Depth Interval (inches)	Nuclide Concentrations (dpm/100 g.)				Total Activity in Interval (dpm)				Total Activity Above Bottom of Interval (dpm)			
	Sr^{90}	Cs^{137}	Ce^{144}	Ru^{106}	Sr^{90}	Cs^{137}	Ce^{144}	Ru^{106}	Sr^{90}	Cs^{137}	Ce^{144}	Ru^{106}
Marlton-8, Collected 8 Aug 1960												
0 - 1/2	366	858	2,573	889	805	1,888	5,661	1,956	805	1,888	5,661	1,956
1/2 - 1	109	240	282	161	446	982	1,153	658	1,251	2,870	6,814	2,614
1 - 2	37.7	47.1	62.9	98.3	362	452	604	944	1,613	3,322	7,418	3,558
2 - 3	13.7	≤ 2.1	19.6	35.7	161	≤ 25	231	420	1,774	3,347	7,649	3,978
3 - 4	6.5	≤ 5.6	5.0	18.8	92	≤ 79	71	266	1,866	3.426	7,720	4,244
4 - 6	2.0	7.2	≤ 6.4	19.3	49	178	≤158	476	1,915	3,604	7,878	4,720
6 - 10	1.0	4.0	≤ 4.8	4.7	52	210	≤252	247	1,967	3,814	8,130	4,967
10 - 12	1.0	4.0	-	-	26	105	-	-	1,993	3,919	-	-
Marlton-8A, Collected 11 Jan 1962												
0 - 1	47.1	96.4	611	120	534	1,094	6,935	1,362	434	1,094	6,935	1,362
1 - 2	8.8	17.8	90	44	80	162	822	400	614	1,256	7,757	1,762
2 - 3	13.9	21.6	178	45	142	221	1,819	462	756	1,477	9,576	2,224
3 - 6	6.3	8.0	137	71	194	244	4,198	2,185	950	1,721	13,774	4,409
6 - 9	2.8	6.3	≤11	34	89	201	≤343	1,077	1,038	1,922	14,117	5,486
9 - 12	6.3	2.9	198	26	201	92	6,292	826	1,239	2,014	20,014	6,312
12 - 18	6.8	4.5	480	50	474	312	33,235	3,476	1,713	2,326	53,644	9,788
18 - 21 1/2	6.6	8.3	710	53	217	274	23,373	1,732	1,930	2,600	77,017	11,520
Marlton-8B, Collected 11 Oct 1962												
0 - 1	34.3	702	3,778	396	162	3,320	17,870	1,873	162	3,320	17,870	1,873
1 - 2	40.8	94.6	82	98	428	993	860	1,027	590	4,313	18,730	2,900
2 - 3	13.5	22.0	47	73	141	230	493	759	731	4,543	19,223	3,659
3 - 6	32.2	11.8	64	66	1,377	505	2,724	2,827	2,108	5,048	21,947	6,486
6 - 9	7.6	3.7	67	58	363	176	3,179	2,774	2,471	5,224	25,126	9.260
9 - 12	8.9	7.1	44	30	308	246	1,515	1,030	2,779	5,470	26,641	10,290
12 - 18	6.8	14.4	113	41	517	1,097	8,609	3,101	3,296	6,567	35,250	13,391
18 - 23 1/2	1.4	10.4	19	38	95	715	1,327	2,599	3,391	7,282	36,577	15,990

TABLE 17 (continued)

RESULTS OF RADIOCHEMICAL ANALYSES OF MARLTON SANDY LOAM

Depth Interval (inches)	Nuclide Concentrations (dpm/100 g.)				Total Activity in Interval (dpm)				Total Activity Above Bottom of Interval (dpm)			
	Sr^{90}	Cs^{137}	Ce^{144}	Ru^{106}	Sr^{90}	Cs^{137}	Ce^{144}	Ru^{106}	Sr^{90}	Cs^{137}	Ce^{144}	Ru^{106}
Marlton-8C, Collected 25 Sep 1963												
0 - 1/2	350	719	2,015	432	1,886	3,875	10,861	2,328	1,886	3,875	10,861	?,328
1/2 - 1	53.9	168	40	133	378	1,179	284	934	2,264	5,505	11,145	3,262
1 - 2	14.3	30.1	50	81	147	309	514	833	2,411	5,363	11,660	4,095
2 - 3	93.5	2.8	36	10.7	1,182	35	459	135	3,593	5,398	12,119	4,230
3 - 4	2.2	5.7	25	5.5	28	73	323	70	3,621	5,471	12,442	4,300
4 - 6	9.3	8.0	70	31	210	181	1,575	703	3,830	5,652	14,017	5,003
6 - 9	7.4	9.5	207	24	256	326	7,121	843	4,086	5,978	21,138	5,846
9 - 12	8.2	18.1	100	10.6	305	671	3,688	393	4,391	6,649	24,826	6,239
12 - 18	22.0	3.8	91	7.1	1,699	297	7,057	548	6,090	6,946	31,883	6,787

TABLE 18

RESULTS OF RADIOCHEMICAL ANALYSES OF LAKEWOOD SAND

Depth Interval (inches)	Nuclide Concentrations (dpm/100 g.)				Total Activity in Interval (dpm)				Total Activity Above Bottom of Interval (dpm)			
	Sr^{90}	Cs^{137}	Ce^{144}	Ru^{106}	Sr^{90}	Cs^{137}	Ce^{144}	Ru^{106}	Sr^{90}	Cs^{137}	Ce^{144}	Ru^{106}
Lakewood-10, Collected 8 Aug 1960												
0 - 1/2	30	318	620	165	121	1,301	2,536	675	121	1,301	2,536	675
1/2 - 1	37	81	129	83	235	512	815	523	356	1,813	3,351	1,198
1 - 2	13.4	6.8	18	47	150	76	196	529	506	1,889	3,548	1,727
2 - 3	20	4.6	14	27	250	58	174	336	756	1,946	3,722	2,064
3 - 4	30	5.0	15	20	377	62	188	248	1,133	2,008	3,910	2,312
4 - 6	8.5	5.7	≤11	9	231	155	≤310	233	1,364	2,163	4,219	2,545
6 - 9	7.1	5.0	33	16	284	200	1,337	652	1,648	2,363	5,556	3,197
9 - 12	5.0	4.3	≤17	13	200	172	≤680	508	1,848	2,535	6,240	3,705
Lakewood-10A, Collected 11 Jan 1962												
0 - 1	12.8	329	822	62	145	3,734	9,330	708	145	3,734	9,330	708
1 - 2	9.1	39	204	58	123	527	2,778	797	268	4,261	12,108	1,505
2 - 3	9.2	11.6	50	75	126	158	677	1,019	394	4,419	12,785	2,524
3 - 6	3.1	13.4	96	69	119	517	3,712	2,651	513	4,936	16,497	5,175
6 - 9	54	5.8	161	71	2,320	248	6,944	3,058	2,833	5,184	23,441	8,233
9 - 12	6.6	2.7	98	39	264	108	3,900	1,537	3,097	5,292	27,342	9,770
12 - 18	3.8	3.8	293	37	264	265	20,620	2,562	3,362	5,557	47,960	12,352
Lakewood-10B, Collected 11 Oct 1962												
0 - 1	120	186	1,406	80	1,242	1,925	14,552	830	1,242	1,925	14,552	830
1 - 2	25	22	145	44	278	245	1,630	494	1,520	2,170	16,181	1,324
2 - 3	6.7	37	27	86	67	371	265	855	1,587	2,541	16,446	2,179
3 - 6	15.2	4.7	84	33	505	156	2,785	1,082	2,092	2,697	19,231	3,261
6 - 9	3.3	3.6	49	24	133	142	1,959	948	2,225	2,840	21,190	4,208
9 - 12	4.3	4.5	147	31	182	191	6,233	1,327	2,407	3,031	27,423	5,535
12 - 18	3.7	11.4	85	36	287	890	6,646	2,812	2,694	3,921	34,069	8,347
18 - 23 1/2	3.2	9.2	58	33	201	583	3,663	2,124	2,895	4,504	37,732	10,471

TABLE 18 (continued)

RESULTS OF RADIOCHEMICAL ANALYSES OF LAKEWOOD SAND

Depth Interval (inches)	Nuclide Concentrations (dpm/100 g.)				Total Activity in Interval (dpm)				Total Activity Above Bottom of Activity (dpm)			
	Sr^{90}	Cs^{137}	Ce^{144}	Ru^{106}	Sr^{90}	Cs^{137}	Ce^{144}	Ru^{106}	Sr^{90}	Cs^{137}	Ce^{144}	Ru^{106}
Lakewood-10C, Collected 25 Sep 1963												
0 - 1/2	72	462	1,193	74	664	4,287	11,071	690	664	4,287	11,071	690
1/2 - 1	33	31	35	67	214	200	231	438	878	4,487	11,302	1,128
1 - 2	11.0	7.5	14	22	131	90	170	257	1,009	4,577	11,472	1,385
2 - 3	7.3	20	43	38	91	252	534	471	1,100	4,829	12,006	1,856
3 - 4	4.2	7.8	20	20	62	116	302	298	1,162	4,946	12,308	2,154
4 - 6	9.8	9.0	60	30	254	233	1,553	774	1,416	5,179	13,861	2,928
6 - 9	24	2.9	21	39	975	120	860	1,589	2,391	5,300	14,721	4,517
9 - 12	4.5	7.8	39	19	170	296	1,479	711	2,561	5,596	16,200	5,228
12 - 16	4.2	32	26	11	217	1,635	1,352	581	2,778	7,231	17,552	5,809
16 - 20	5.3	2.6	34	13	259	126	1,676	617	3,037	7,357	19,228	6,426

TABLE 19

COMPARISON OF CUMULATIVE DEPOSITS OF STRONTIUM-90 AND CESIUM-137 IN THE MARLTON AND LAKEWOOD SOILS WITH TOTAL DEPOSITION AT WESTWOOD, NEW JERSEY

Sample	Sr^{90} Deposit (mc/mi^2) 0 - 12"	Sr^{90} Deposit (mc/mi^2) Entire Core	Cs^{137} Deposit (mc/mi^2) 0 - 12"	Cs^{137} Deposit (mc/mi^2) Entire Core
Collection Date: 8 August 1960				
Core 8	73	73	140	140
Core 10	65	65	90	90
Average	69	69	115	115
Precipitation	84		142	
Collection Date: 11 January 1962				
Core 8A	44	69	72	93
Core 10A	111	118	190	201
Average	78	93	131	147
Precipitation	89		154	
Collection Date: 11 October 1962				
Core 8B	100	123	197	258
Core 10B	86	100	108	158
Average	93	112	152	208
Precipitation	115		193	
Collection Date: 25 September 1963				
Core 8C	154	215	239	250
Core 10C	93	110	201	262
Average	123	162	220	256
Precipitation	176		289	

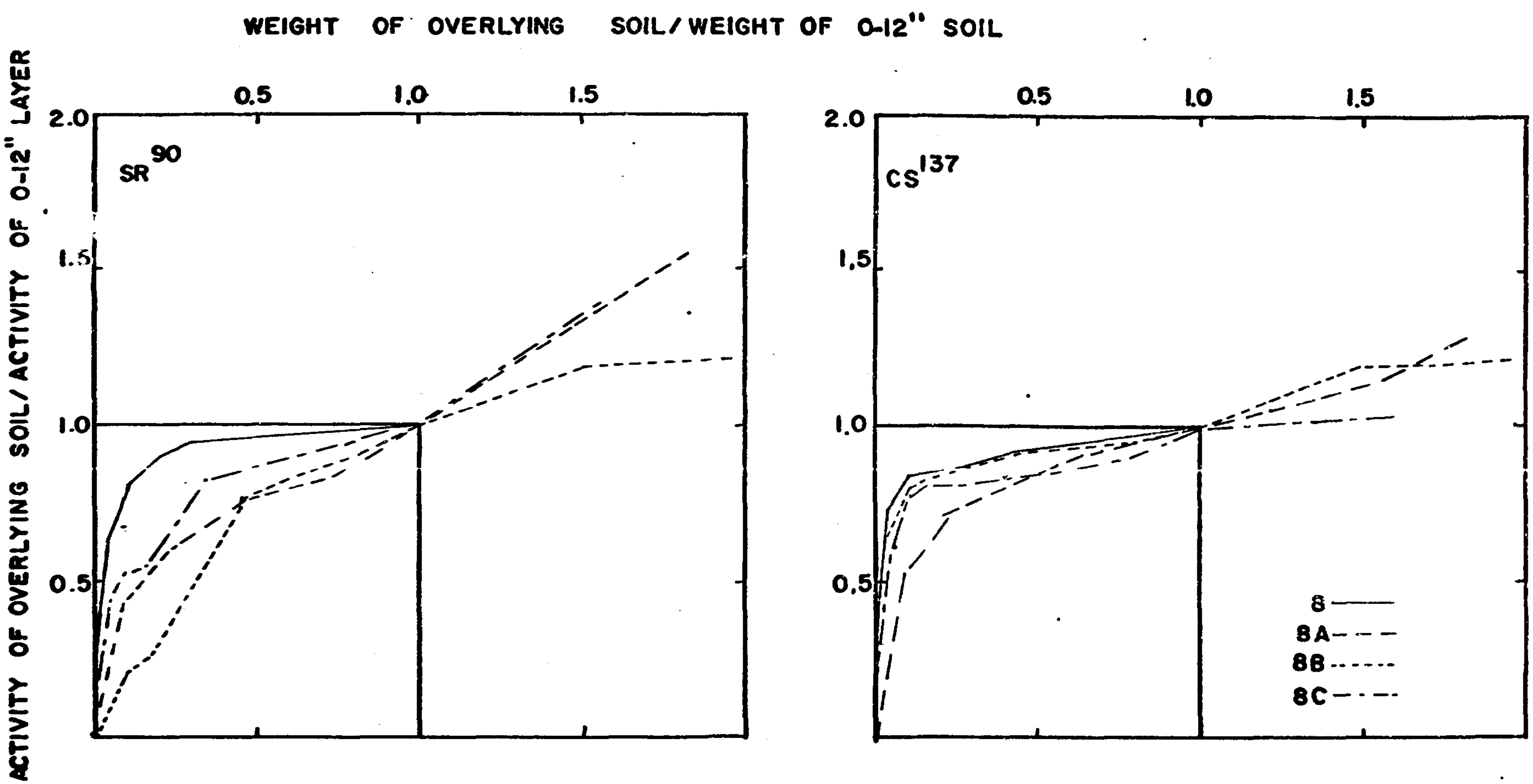


FIGURE 85. VARIATIONS IN PENETRATION OF MARLTON SANDY LOAM (CORES 8, 8A, 8B, 8C) BY Sr^{90} AND Cs^{137}

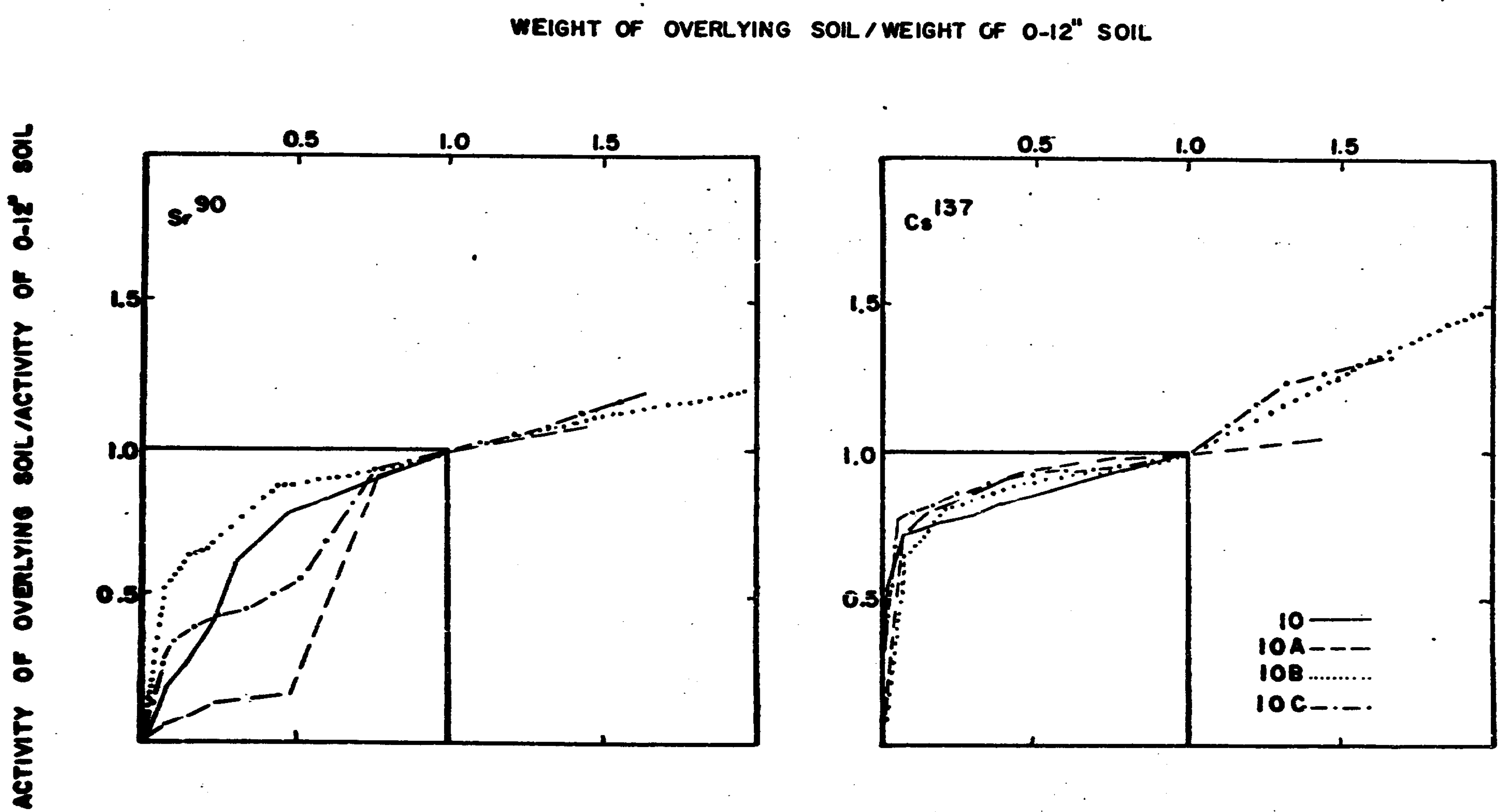


FIGURE 86. VARIATIONS IN PENETRATION OF LAKEWOOD SAND (10, 10A, 10B, 10C) BY Sr^{90} AND Cs^{137}

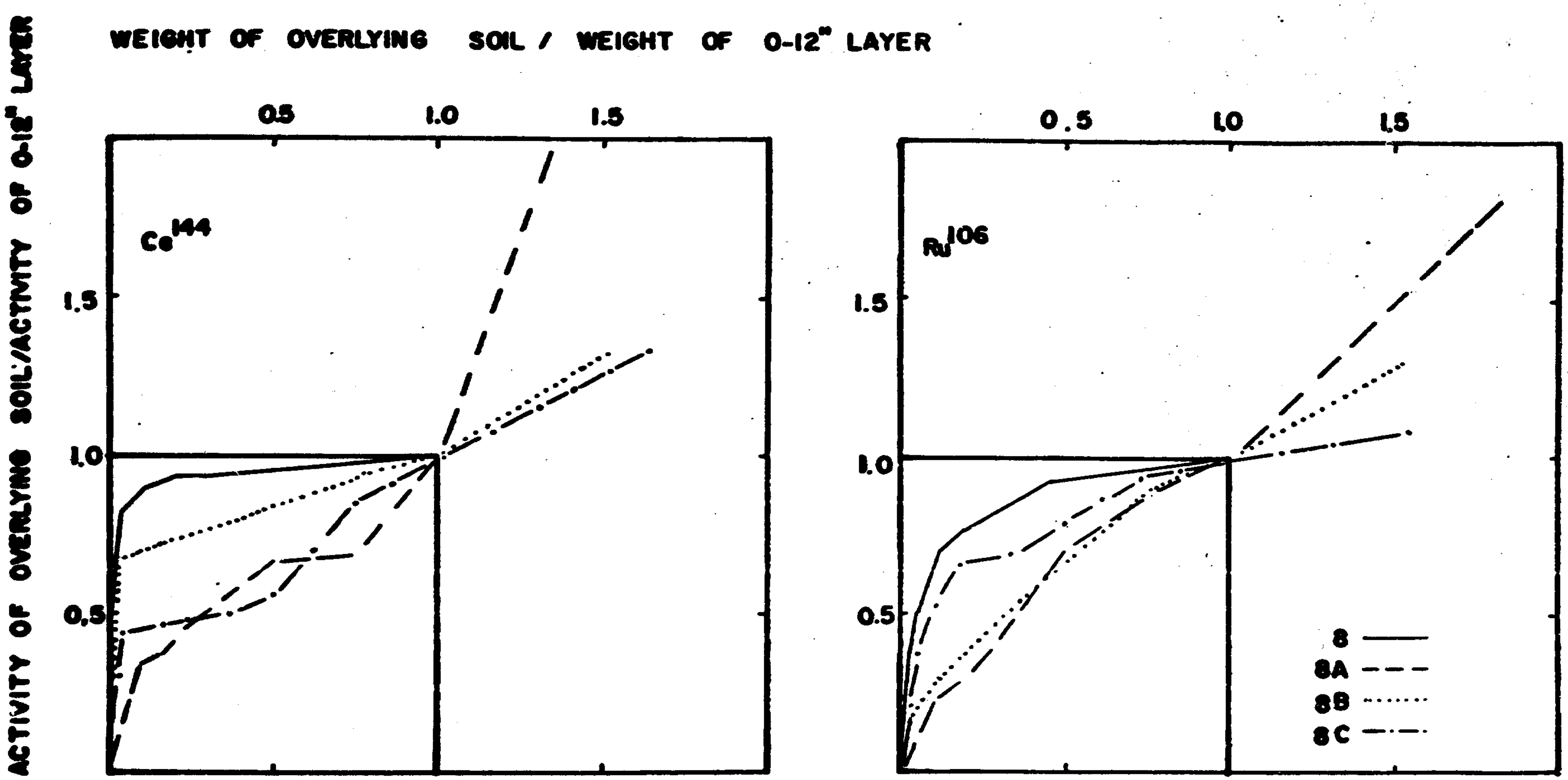


FIGURE 87. VARIATIONS IN PENETRATION OF MARLTON SANDY LOAM (CORES 8, 8A, 8B, 8C) BY Ce^{144} AND Ru^{106}

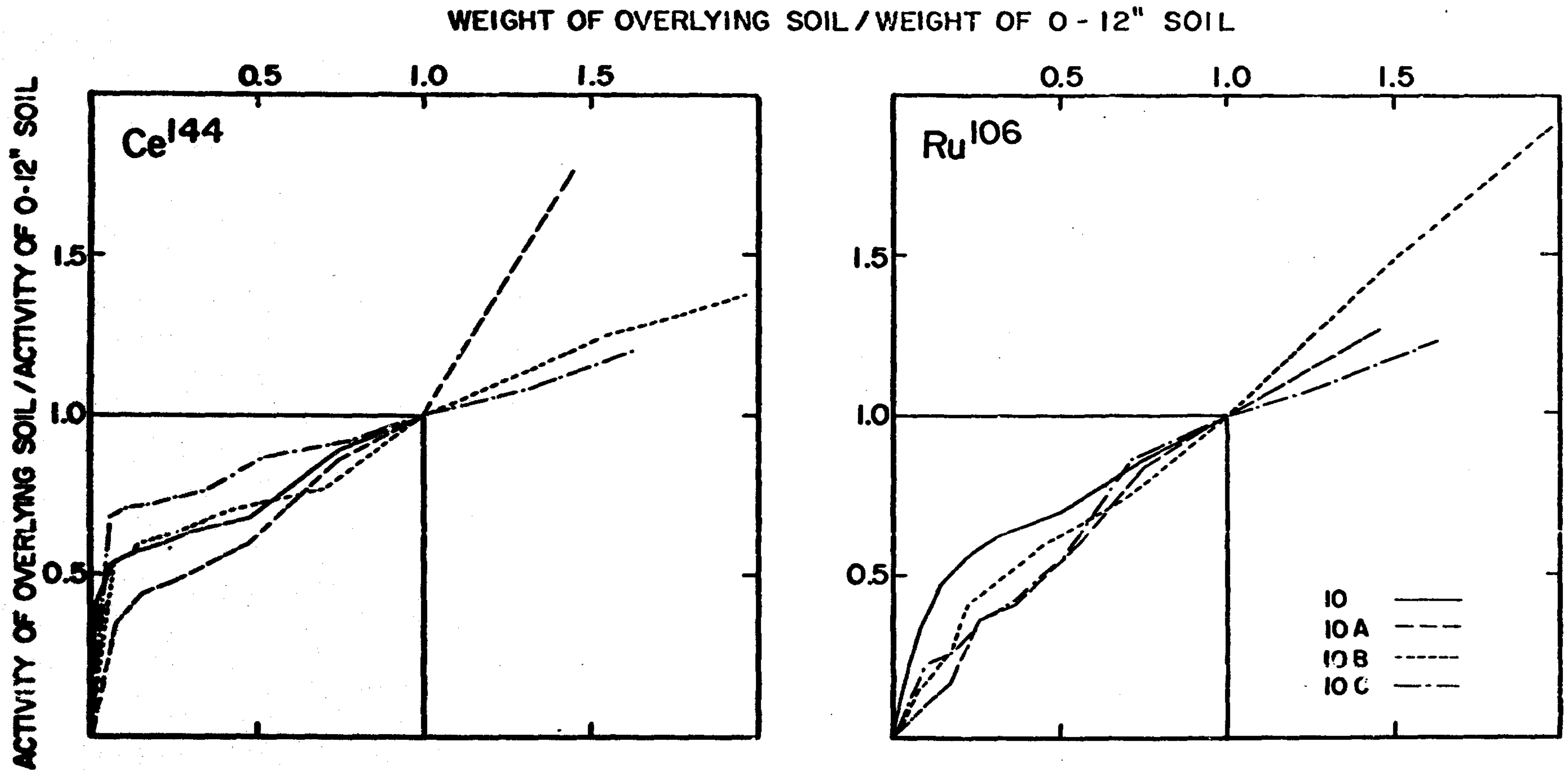


FIGURE 88. VARIATIONS IN PENETRATION OF LAKEWOOD SAND (Cores 10, 10A, 10B, 10C) BY Ce-144 AND Ru-106

CHAPTER 10. SUMMARY

The progress made on Project Stardust during September, 1963 to September, 1965 is reviewed in this report. Changes made in the techniques of sample collection and analyses since the publication of DASA-1309, the Ninth Quarterly Report on Project Stardust, are described. Results of measurements of filter and gas samples of stratospheric air are discussed, progress in the development of the Stardust model of atmospheric transfer and rainout is summarized, and data on carbon-14 in ground-level air and on fission products in New Jersey soils are presented.

The Stardust Sampling Program

Beginning in August, 1963 whole air samples have been collected during most Stardust sampling missions flown in the Northern Hemisphere. Filter samples have been collected on all missions flown over the entire range of latitude of the Stardust sampling corridor, from 75°N to 55°S. Since 1963 there has been a gradual reduction in the number of Stardust sampling missions flown, and by mid-1965 the sampling rate was less than half of the sampling rate of mid-1963. By early 1964 the RB-57F had replaced the WU-2 as the main sampling vehicle on Stardust missions.

The Stardust Analytical Program

During 1963 to 1965 Stardust filter samples were analysed for a series of fission products, activation products, and natural radionuclides, including several cosmic-ray products. As activities decreased, and as debris from different injections became rather well mixed, most of these nuclides were eliminated from the scheme of analysis. By late 1965 these samples were being analysed only for strontium-90, plutonium-238, 239, cadmium-109, manganese-54 and, occasionally, lead-210 and, polonium-210.

Aliquots of the Stardust gas samples were analysed for their carbon dioxide content by means of an isotope dilution technique, and the separated carbon dioxide

fractions were analyzed for carbon-14.

The Stratospheric Distribution of Weapons Debris

Very high concentrations of radioactive debris were found in the stratosphere during early 1963 as a result of injections during the 1961 and 1962 nuclear weapons tests series. These concentrations decreased rapidly during 1963 to 1965 as a result of fallout and radioactive decay. The total fission product activities encountered in the northern polar stratosphere were about ten times as high as those encountered at equivalent locations in the southern polar stratosphere in late 1963, but by late 1965 they were only twice as high.

Strontium-90 concentrations at 65 thousand feet at 65°N and 25°N decreased by about a factor of nine between early 1963 and late 1965, from about 2250 dpm/1000 SCF to about 250 dpm/1000 SCF. At 65 thousand feet at 35°-40°S strontium-90 concentrations increased by more than a factor of three during the second half of 1963, from about 110 dpm/1000 SCF to about 360 dpm/1000 SCF, as radioactive debris moved from the tropical stratosphere into the southern polar stratosphere, but by the second half of 1965 the strontium-90 concentration at this location had decreased to about 100 dpm/1000 SCF. There appeared to be an acceleration in the rate of movement of debris into the southern polar stratosphere in May-September 1963, May-June 1964, and June-September 1965. The vertical profiles of strontium-90 activity, in the Northern Hemisphere at least, have contained a layer of maximum concentration within the region of the stratosphere sampled by aircraft or balloons. This maximum has been found at all latitudes, but its height increases from the pole toward the equator. It has been found between 50 and 70 thousand feet at 65°-70°N, between 60 and 85 thousand feet at 30°-35°N, and between 65 and 90 thousand feet at 10°N. The decrease in the stratospheric strontium-90 burden during 1963-1965, from 6.5 ± 1.5 megacuries in early 1963, to 3.0 ± 0.5 megacuries by early 1964, and to 0.9 ± 0.4 megacurie by mid-1965, corresponds to a stratospheric residence half-time of about 10 months for this particulate debris.

During 1961-1965 the activity ratios Cs^{137}/Sr^{90} (1.56 ± 0.24) and $Pu^{239,240}/Sr^{90}$ (0.016 ± 0.005) generally showed little variation with location or time of sample collection (except for a low $Pu^{239,240}/Sr^{90}$ ratio typical of samples collected in the tropical stratosphere during May-December 1962), suggesting that these nuclides have had rates of injection and of fallout which have borne a relatively constant ratio to the rates of injection and fallout of strontium-90.

Manganese-54 and several other products of neutron activation injected into the stratosphere by the 1961 and 1962 weapons tests also showed rates of transfer within the stratosphere and rates of fallout to the troposphere and to the ground similar to those of strontium-90 during 1963 to 1965. Like strontium-90, manganese-54 displayed a stratospheric residence half-time of 10 months during 1963 to 1965, with its stratospheric burden, corrected for decay to 31 December 1962, decreasing from about 24 megacuries in early 1963, to about 10 megacuries in early 1964, and to 3 megacuries in mid-1965. Iron-55, another activation product injected by the 1961 and 1962 tests, behaved in a similar manner to manganese-54. The activity ratio Fe^{55}/Mn^{54} (corrected for decay to 31 December 1962) remained fairly constant at 2.4 ± 0.4 in Stardust samples throughout 1963 to 1965. Because of the relatively short half-life of antimony-124, changes in the stratospheric distribution of this activation product could be followed only into early 1964. It was determined, however, that relatively little of this nuclide entered the southern polar stratosphere during mid-1963. Probably most of the debris that did enter the southern polar stratosphere during mid-1963 had been produced by the 1961 USSR weapons tests and the 1962 U.S. low latitude tests, rather than by the 1962 USSR high latitude tests. Yttrium-88, which was produced by one or more tests in late 1962, did enter the southern polar stratosphere during mid-1963, but this nuclide may have been injected into the stratosphere at low latitudes. Artificial sodium-22 produced by the 1961 and 1962 test series apparently entered the southern polar stratosphere by mid-1964, but its concentrations were low and not sufficiently higher than the natural concentrations

of this nuclide to allow quantitative interpretation of the data.

A small amount of cadmium-109 may have been produced by the late 1962 USSR weapons tests, but by mid-1963 this nuclide was not measurable in Stardust filter samples. Cadmium-109 produced by the Starfish Prime high altitude event was first intercepted by Stardust sampling missions in the southern polar stratosphere towards the end of 1963, and in the northern polar stratosphere at about the beginning of 1964. During 1964 the cadmium-109 concentrations in the lower polar stratosphere were far higher in the Southern than in the Northern Hemisphere, but by mid-1965 there was little difference in concentration between the two hemispheres. Moreover, by then there appeared to be a fairly uniform vertical distribution of cadmium-109 between the upper stratosphere and levels as low as 55 thousand feet in the lower stratosphere. It is estimated that about 70 kilocuries of cadmium-109 (corrected for decay to 9 July 1962) were present in the stratosphere in 1964, and that by mid-1965 about 25 kilocuries, half of the remaining burden, were present between the tropopause and the 50 mb level (68 thousand feet).

Excess carbon-14 produced by the 1961 and 1962 nuclear weapons tests has exhibited horizontal and vertical concentration profiles in the stratosphere which differ from those exhibited by particulate debris from these tests, and the differences are attributed largely to the effects of particle settling on the particulate debris. The burden of excess carbon-14 in the stratosphere of the Southern Hemisphere remained between 5×10^{27} and 6×10^{27} atoms during 1963 to 1965, but the burden in the stratosphere of the Northern Hemisphere decreased from about 30×10^{27} atoms in early 1963 to about 15×10^{27} atoms in mid-1964 and to about 10×10^{27} atoms in mid-1965. The residence half-time of excess carbon-14 in the stratosphere averaged about 18 months during 1963 to 1965, but increased with the passage of time.

<u>The Stratospheric Distribution of SNAP-9A Plutonium-238</u>

Plutonium-238 injected into the upper stratosphere on 21 April 1964 by the burn-up of a SNAP-9A power source was first intercepted by Stardust sampling missions

in May 1965 in the southern polar stratosphere. Increasing amounts of SNAP-9A plutonium-238 were intercepted in the lower northern polar stratosphere during the second half of 1965, and by December, 1965 significant concentrations of this nuclide were found in that region. The plutonium-238 data demonstrate that the mechanisms of atmospheric transport which cause the movement of radioactive debris from the upper into the lower stratosphere are capable of transferring significant quantities of the debris through virtually the entire thickness of the lower stratosphere (from above the 40 millibar level down to below the 150 mb level at least) over the course of a few months. Such rapid transfer is more readily explained in terms of eddy diffusion than in terms of mean advective transport within the stratosphere. An estimate of the stratospheric burden of SNAP-9A plutonium-238 during September-December 1965, based on Stardust data and USAEC balloon data, gives 7.5 kilocuries, less than half of the 17 kilocuries injected. Only 2.4 kilocuries were calculated to be present in the lower stratosphere below the 40 millibar level (72 thousand feet), and most of the discrepancy between the actual injection and the calculated burden probably results from our underestimating the concentrations in the upper stratosphere.

The Stratospheric Distribution of Natural Radioactivity

The stratospheric distributions of the cosmic ray products beryllium-7 and phosphorus-32 during 1963 to 1965 were generally similar to the theoretical distribution in a stagnant atmosphere. In the northern polar stratosphere, however, in the vicinity of the Stardust sampling corridor the concentrations of these two nuclides apparently decreased between December, 1963 and February, 1964 as a result of a seasonal shift in the stratospheric circulation and the influx of air into that region from lower latitudes.

Lead-210 concentrations in filter samples collected during 1957 to 1959 indicate a stratospheric distribution of lead-210 with a layer of maximum concentration in the lower stratosphere, at a height of about 45 thousand feet in the polar

stratosphere but sloping upward toward the equator to a height of 60 thousand feet in the tropical stratosphere. Measurements of the Po^{210}/Pb^{210} activity ratio in samples collected during December, 1964 indicate that this ratio is well below 1.0 in the upper troposphere, but that in the lower stratosphere it is approximately equal to 1.0

Progress on the Stardust Transfer and Rainout Model

Work continued on the development of a numerical model in two dimensions which will be capable of simulating diffusion, transport, particle fall velocity and tropospheric rainout. The model incorporates a general anisotropic diffusion process. Rainout is simulated by fractional removal of debris at selected "rain formation" levels; the fraction removed at any latitude is proportional to the observed rainfall at that latitude. In most versions of the model thus far tested the transport terms have been neglected and an attempt has been made to reproduce available data with a diffusion - rainout - settling velocity model.

The parameters of the model have been varied in an attempt to reproduce quantitatively the known behavior of tungsten-185 injections in equatorial regions and, at least qualitatively, the behavior of high latitude stratospheric injections. Three types of models have been described; they are characterized respectively by the principal diffusion axis being (a) along a horizontal surface, (b) along surfaces of constant potential temperature, and (c) approximately along surfaces of constant potential vorticity. All three types can reproduce the latitudinal variation of rainout in a satisfactory manner, but only the third can reproduce the observed pattern of concentration in the stratosphere.

Carbon-14 in Ground Level Air

The concentration of carbon-14 in carbon dioxide of ground level air in northern New Jersey has decreased since 1964. The concentration of excess carbon-14

from nuclear weapons tests ranged between 10 and 25 percent of the concentration of natural carbon-14 during 1960 and 1961, but as a result of the 1961 and 1962 test series it rose to over 40 percent of the natural level in mid-1962, and to over 90 percent in mid-1963 and mid-1964. In mid-1965, however, it reached only 75 percent of the natural level. Each year since 1960 the carbon-14 concentration in ground level carbon dioxide has reached a maximum during the summer and a minimum during the winter.

Fission Products in New Jersey Soils

Measurements have been made of the vertical distributions of the fission products strontium-90, cesium-137, cerium-144 and ruthenium-109 in cores of two New Jersey soils, the Marlton sandy loam and the Lakewood sand. Cores were collected in August, 1960, January, 1962, October, 1962 and September, 1963 to permit study of changes with time in these vertical distributions. The results of the analyses are not of the best quality, but they appear adequate to permit the drawing of a few general conclusions. There is some evidence that activity profiles in the January, 1962 core were flatter than those in the August, 1960 core, probably as a result of downward migration of the radionuclides. The activity profiles in the October, 1962 and September, 1963 cores were steeper than those in the earlier cores, probably because of the increased rate of deposition of fallout on the surface following the 1961 and 1962 weapons tests. By averaging total activities of strontium-90 and cesium-137 in those Marlton and Lakewood cores which were collected together, it is possible to obtain estimates of the rate of deposition of fallout in New Jersey which agree reasonably well with estimates based on measurements of fallout in precipitation at Westwood, New Jersey.

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13. ABSTRACT

Work performed on Project Stardust during the two year period, September 1963 to September 1965 is reviewed. Changes in the sampling and analytical programs are summarized. The stratospheric distributions of radioactive debris from nuclear weapon tests are described. Nuclides described include strontium-90, cesium-137, plutonium-239, manganese-54, iron-55, antimony-124, yttrium-88, sodium-22, cadmium-109, silver-110 and carbon-14. The stratospheric distributions of plutonium-238 from a SNAP-9A burnup, of several cosmic ray products and of lead-210 and plutonium-210 are discussed. Progress in the design of the Stardust numerical model of transfer and rainout of stratospheric radioactive materials is summarized.

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